Tunneling resonances in quantum dots: Coulomb interaction modifies the width

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Single-electron tunneling through a zero-dimensional state in an asymmetric double-barrier resonant-tunneling structure is studied. The broadening of steps in the *I-V* characteristics is found to strongly depend on the polarity of the applied bias voltage. Based on a qualitative picture for the finite-lifetime broadening of the quantum dot states and a quantitative comparison of the experimental data with a nonequilibrium transport theory, we identify this polarity dependence as a clear signature of Coulomb interaction.

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Single-electron tunneling through zero-dimensional states has been observed in a wide variety of systems, including metallic islands, lateral quantum dots in gated semiconductor devices, vertical dots in double-barrier resonant tunneling structures, and molecular systems such as carbon nanotubes.[1] The (*I-V*) characteristics has the shape of a staircase, in which each step is associated with the opening of a new transport channel through the system. Many features observed in the transport measurements[2–6] can be explained either within a single-particle picture for noninteracting electrons or by the orthodox theory of sequential tunneling[7–9] valid for weak dot-lead tunnel coupling. This is no longer the case for interaction effects on transport beyond the weak-tunneling limit. A famous example is the zero-bias anomaly of Kondo-assisted tunneling.[10,11]

In this work, we report on a clear signature of Coulomb interaction beyond weak tunneling that is achieved under much less stringent experimental conditions than required for the Kondo effect to occur. This signature is contained in the width of the first step of the *I-V* characteristics. We observe that the width strongly depends on the polarity of the applied bias voltage. This behavior can neither be explained within a single-particle picture nor by sequential-tunneling theory.[7,8,12] Due to Coulomb interaction, the finite-lifetime broadening of the dot levels becomes energy dependent. As a consequence, the values for the broadening at the two considered steps of opposite polarity can differ by up to a factor of two for strongly asymmetric coupling strengths of the two tunnel barriers. We use the results of a diagrammatic real-time transport theory[13] that includes the above described physics to find reasonable quantitative agreement with the experimental data.

The experiment was performed with a highly asymmetric double-barrier resonant-tunneling device grown by molecular beam epitaxy on an n^+-type GaAs substrate. An undoped 10-nm-wide GaAs quantum well is sandwiched between 5- and 8-nm-thick $Al_{0.3}Ga_{0.7}As$ tunneling barriers separated from highly doped GaAs contacts (Si-doped with $n_{Si}=4\times10^{17}$ cm^{-3}) by 7-nm-thick undoped GaAs spacer layers. The sample was fabricated as a pillar of 2 μm diameter. Two-terminal dc measurements of the *I-V* characteristics were performed in a dilution refrigerator at temperatures between 20 mK and 1 K. The studied GaAs quantum well embedded between two AlGaAs barriers can be viewed as a two-dimensional system with the edges and residual impurities confining the lateral electron motion and thus forming dots. We determine the parameters of these *natural* quantum dots by detailed magnetic-field-dependent spectroscopic measurements.[15] Here, we investigate tunneling through the energetically lowest state of a dot, at the energy $E_0=33$ meV and with a lateral extent of 10 nm corresponding to a single-particle level separation of the order of 30 meV.

The system considered is sketched in Fig. 1(a). Electrons tunnel from the heavily doped emitter through the spin-degenerate quantum-dot level embedded inside the quantum well. The level comes to resonance with the emitter's electro-chemical potential at a finite bias voltage, indicated by a step in the current, as shown in Fig. 1(b).

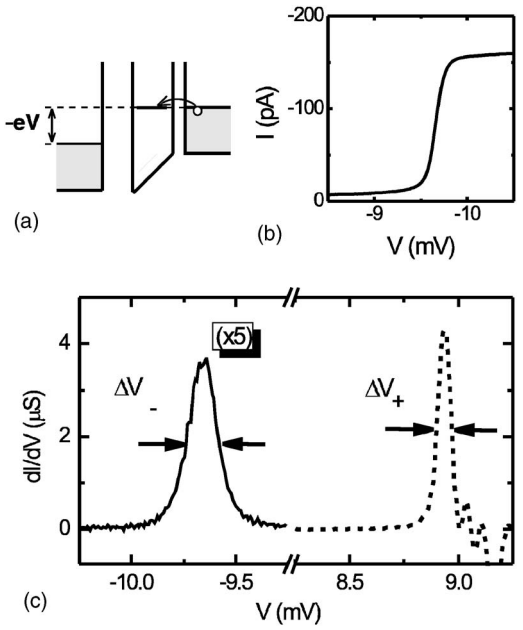

FIG. 1. (a) Schematic energy diagram of the asymmetric double-barrier device under finite bias. (b) First current step in negative bias direction. (c) Comparison of the full width half maximum (FWHM) value of the differential-conductance peaks for both bias polarities at base temperature ($T=20$ mK).

Coulomb interaction prevents double occupation of the level for the considered range of bias voltage. This gives rise to charging effects that are differently important for the two polarities. The bottleneck of transport is provided by the thicker tunnel barrier. For $V<0$, the "charging direction," as sketched in Fig. 1(a), the dot is predominantly singly occupied, and, therefore, the two spin channels effectively block each other. This reduces the current-step height to one-half of the value for the opposite polarity, $V>0$, the "noncharging direction," for which the dot is predominantly empty.[4–6,8,9,12] This is well understood within the sequential-tunneling picture.

The focus of this paper, however, is the finite broadening of the step edge. This broadening can be measured as the full width at half maximum (FWHM) value of the differential conductance (dI/dV) peaks associated with the current steps. Figure 1(c) shows our experimental low-temperature differential conductance peaks for both bias polarities. As a result, the width $\Delta V_-=152\ \mu$V for negative bias has roughly twice the value than that of $\Delta V_+=71\ \mu$V. In previous experiments the resonance width in asymmetric structures has been already studied and a polarity dependence has been observed, but their explanations referred to polarity-dependent leverage factors[3] or internal saturation processes.[5]

The width of the current step edge at low temperature reflects the finite-lifetime broadening of the zero-dimensional state due to tunneling in and out of the dot. The golden-rule rates for an electron tunneling in and out of the dot are given by $\Gamma^+(\omega)=\Sigma_{r=\mathrm{L,R}}\Gamma_r f_r(\omega)$ and $\Gamma^-(\omega)=\Sigma_{r=\mathrm{L,R}}\Gamma_r[1-f_r(\omega)]$, respectively. The tunnel-coupling strength is characterized by the constant $\Gamma_r=2\pi\nu_r|t_r|^2$, where ν_r is the density of states in lead r, and t_r is the tunneling amplitude, and $f_r(\omega)=f(\omega-\mu_r)$ is the Fermi function of lead r with electrochemical potential μ_r. We note that the presence of strong Coulomb interaction introduces an asymmetry between tunneling-in and tunneling-out processes; while for an empty dot there are two possibilities to choose the spin state of the incoming electron, the spin state of an electron leaving the dot is fixed. It is, therefore, the combination $2\Gamma^+(\omega)+\Gamma^-(\omega)$, i.e.,

$$\sum_r \Gamma_r[1+f_r(\omega)], \tag{1}$$

that determines the finite-lifetime broadening. This expression is energy dependent, i.e., it depends on the relative position of the relevant transport channels to the Fermi energy of the leads. At low temperature, the broadening probed by the electrons within the transport window set by the Fermi energies of emitter and collector is either $2\Gamma_\mathrm{L}+\Gamma_\mathrm{R}$ or $\Gamma_\mathrm{L}+2\Gamma_\mathrm{R}$, depending on whether the left lead serves as emitter or collector. This qualitatively explains the polarity-dependent step width. It is an interaction effect since in the absence of Coulomb interaction the asymmetry between tunneling-in and tunneling-out processes is lifted by processes involving double occupancy of the dot, and the finite-lifetime broadening is energy independent, given by $\Gamma_\mathrm{L}+\Gamma_\mathrm{R}$.

For a more quantitative analysis we employ a real-time diagrammatic technique[13] that was developed for a systematic investigation of higher-order tunneling processes in nonequilibrium transport through an interacting quantum dot. In particular, we make use of the results for the current obtained within a nonperturbative resummation scheme, the so-called resonant-tunneling approximation for transport through a single (spin-degenerate) level in the presence of strong Coulomb interaction such that double occupancy is prohibited.[14] The technique, the approximation scheme, and the steps of the calculation are presented in Ref. 13. The result for the current is

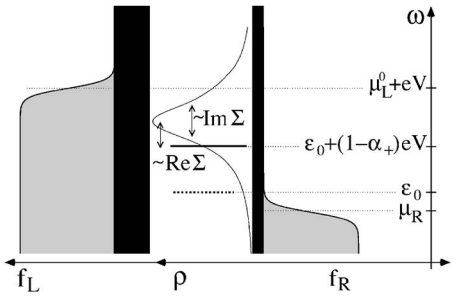

FIG. 2. Sketch of our device in a noncharging direction. The electronic structure of the dot is given by the spectral density $\rho(\omega)$, which is shifted and broadened with respect to the bare level $\varepsilon_0+(1-\alpha_+)eV$ (solid line) as reflected in the real part and imaginary part of the self-energy Σ, respectively.

$$I=\frac{e}{h}\int_{-\infty}^{\infty}d\omega\frac{2\Gamma_\mathrm{L}\Gamma_\mathrm{R}[f_\mathrm{L}(\omega)-f_\mathrm{R}(\omega)]}{[\omega-\varepsilon-\mathrm{Re}\,\Sigma(\omega)]^2+[\mathrm{Im}\,\Sigma(\omega)]^2}, \tag{2}$$

where ε is the dot level energy [with the appropriate incorporation of a bias voltage influencing level energy $\varepsilon=\varepsilon_0+(1-\alpha_+)eV$ and Fermi energies of the leads; $\mu_\mathrm{R}=\mathrm{const}<\varepsilon_0$, $\mu_\mathrm{L}=\mu_\mathrm{L}^0+eV$ for the noncharging direction] and the self-energy (see Fig. 2),

$$\Sigma(\omega)=\sum_r\frac{\Gamma_r}{2\pi}\int d\omega'\frac{1+f_r(\omega')}{\omega-\omega'+i0^+}. \tag{3}$$

Evaluating the integral leads to

$$\mathrm{Re}\,\Sigma(\omega)=\sum_r\frac{\Gamma_r}{2\pi}\left[\ln\left(\frac{\beta E_C}{2\pi}\right)-\mathrm{Re}\,\psi\left(\frac{1}{2}+i\frac{\beta(\omega-\mu_r)}{2\pi}\right)\right]$$

and

$$\mathrm{Im}\,\Sigma(\omega)=-\sum_r\frac{\Gamma_r}{2}[1+f_r(\omega)]. \tag{4}$$

The real part is weakly dependent on a high-energy cutoff E_C given by the smaller of the charging energy for double occupancy or the bandwidth of the leads. In the imaginary part, we recover the structure of the finite-lifetime broadening as postulated in the qualitative discussion above. Deep in the Kondo regime the approximation above is no longer valid. The two spin channels become independent from each other[10] and $\mathrm{Im}\,\Sigma(\omega)=-\Sigma_r\Gamma_r/2$.

We remark that for the *step height* the well-known (sequential-tunneling) result $\Delta I_+=(2e/\hbar)\Gamma_\mathrm{L}\Gamma_\mathrm{R}/(2\Gamma_\mathrm{L}+\Gamma_\mathrm{R})$

for the noncharging direction (and $L \leftrightarrow R$ for the charging direction) is reproduced.

The focus of this paper, however, is on the *width of the step edge*. At *low temperature*, Eq. (2) simplifies to

$$I_+ = \frac{e}{h}\int_{\mu_R}^{\mu_L^0+eV} d\omega \frac{2\Gamma_L\Gamma_R}{[\omega-\varepsilon-\mathrm{Re}\,\Sigma(\omega)]^2+\left[\Gamma_L+\frac{\Gamma_R}{2}\right]^2} \quad (5)$$

with $\varepsilon=\varepsilon_0+(1-\alpha_+)eV$ and α_+ being the leverage factor for positive bias (and $\alpha_-=1-\alpha_+$ being the leverage factor for negative bias), denoting the voltage drop over the left barrier. Neglecting the real part of the self-energy for the moment, we find that the differential conductance as a function of V is a Lorentzian with a FWHM of

$$\alpha_+ e\Delta V_+ = 2\Gamma_L + \Gamma_R \quad \text{and} \quad \alpha_- e\Delta V_- = \Gamma_L + 2\Gamma_R.$$

For $\alpha_+ \approx \alpha_-$ and strongly asymmetric tunnel-coupling strengths, $\Gamma_L \ll \Gamma_R$, we get the relation $\Delta V^- \approx 2\Delta V^+$.

At *high temperature* we find for the FWHM $e\Delta V_\pm = 3.525 k_B T/\alpha_\pm + e\Delta V_\pm^0$, i.e., the temperature broadening of the Fermi function. As predicted from sequential-tunneling theory[7] the width increases linearly with temperature. The constant term $\Delta V_\pm^0$ is of the order of Γ, and, in general, also polarity dependent.[18]

For a detailed comparison between theory and experiment we need first to determine the system parameters. The factor α determining the bare level shift with bias voltage is gained from the linear high-temperature dependence of $\Delta V_\pm(T)$ as $\alpha_+=0.53$ and $\alpha_-=0.5$ so that $\alpha_+ + \alpha_- \approx 1$. The coupling constants $\Gamma_{L/R}$ could, in principle, both be determined from the current steps $\Delta I_\pm$. For strong asymmetry, however, as is the case here, the maximum current is limited entirely by the bottleneck of the smaller coupling Γ_L. As a consequence, the step heights only fix $\Gamma_L = \Delta I^+\hbar/(2e) = 2\Delta I^-\hbar/(2e) = 0.64\ \mu$eV.

Considering the results derived above for zero temperature FWHM, we note that this width is essentially the sum of different couplings and, hence, is dominated by the larger coupling Γ_R. Therefore we can find $\Gamma_R = (1-\alpha^+)e\Delta V^+ \approx (1-\alpha^-)\Delta V^-/2$. In fact, we gain better accuracy by fitting the full dI/dV peak for charging polarity and lowest temperature to pinpoint $\Gamma_R = 40\ \mu$eV. The high-energy cutoff $E_C = 30$ meV is given by the bandwidth, i.e., the value of the Fermi energy, being of the same magnitude as the charging energy.

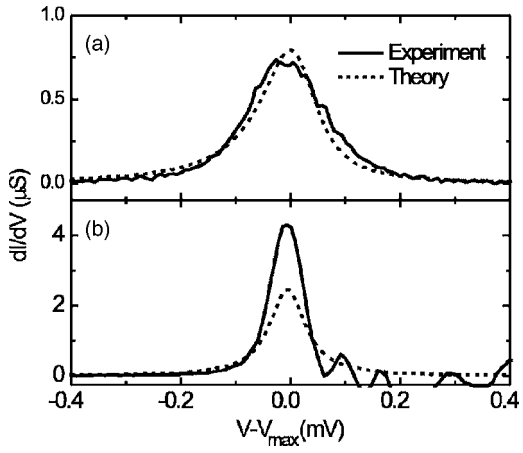

FIG. 3. Experimental (solid lines) and theoretical (dashed lines) differential conductance for the (a) charging and (b) noncharging direction at base temperature ($T=20$ mK).

Figure 3 shows the comparison between experimental data and theoretical calculations for the differential conductance. For the charging direction (negative bias), Fig. 3(a), we find a good agreement between experiment and theory for both the resonance width and amplitude. In the noncharging direction (positive bias), Fig. 3(b), the peak width is reduced by about a factor of two for both experiment and theory. The experimental data show some extra features. First there is an oscillatory fine structure on the positive-voltage side of the resonance attributed to the fluctuations of the local density of states of the emitter (see, e.g., Ref. 16). Moreover, we observe an enhanced resonance amplitude as compared to the theoretical calculation. This effect may be related to an additional many-body phenomenon at the Fermi edge.[17]

The temperature dependence of the resonance width in the broad range between 20 mK and 400 mK is shown in Fig. 4(a). The polarity dependence of the width is clearly visible. At low temperatures experimental and theoretical data nicely match for the different widths at base temperature and for the different saturation temperatures. For temperatures above 200 mK the width increases linearly with $e\alpha_\pm\Delta V_\pm = 3.525 k_B T + \mathcal{O}(\Gamma)$. This experimental result is again adequately reproduced by theory.

Further support for our explanation of the polarity dependence as an interaction effect is given by the magnetic-field dependence. In Fig. 4(b) the measured resonance widths at base temperature are shown as function of the magnetic field

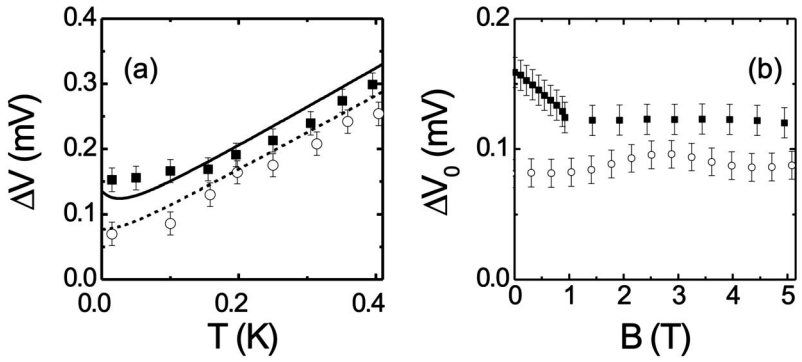

FIG. 4. (a) Temperature dependence of the resonance width ΔV at $B=0$ T for both polarities, open circles $V>0$, filled squares $V<0$, continuous lines theoretical simulations. (b) Resonance width ΔV at base temperature ($T=20$ mK) as a function of B field in direction $B \| I$ for both polarities of applied voltage.

applied parallel to the current. The data reveal that polarity dependence is strongly reduced by a Zeeman splitting (on a scale of ≈ 1 T, which is of the order of the magnitude of the coupling). In this case, only one spin state contributes to transport, and the system is equivalent to a noninteracting one, for which theory predicts $\mathrm{Im}\,\Sigma(\omega)=-\Sigma_r\Gamma_r/2$ and, thus, a low-temperature width $\alpha_\pm e\Delta V_\pm=\Gamma_\mathrm{L}+\Gamma_\mathrm{R}$ independent of the polarity. This is in accordance with the trend seen in the experimental data. The difference between the polarities remaining for a high magnetic field and the slight oscillations with the field point to field effects going beyond our simple picture.

The main discrepancies remaining between experimental data and theoretical simulation are consistent with the expected range of accuracy. The main source of experimental errors are systematic fluctuations in the current, in particular the local density of states fluctuations prominent in noncharging direction which become even more pronounced at high temperatures. This artifact particularly limits the precision to which we can determine the current step height ΔI_+.

We study the width of the current step of single-electron tunneling through a zero-dimensional state in a double-barrier resonant tunneling structure and find that it is strongly polarity dependent. This is interpreted as a clear signature of Coulomb interaction. The latter introduces an asymmetry between possible tunneling-in and tunneling-out processes, which gives rise to an energy-dependent finite-lifetime broadening. The bias-polarity dependent step width, observed in our experiment, where strengths of the tunnel coupling between dot and the two leads were highly asymmetric, can be simulated with reasonable agreement by applying a diagrammatic nonequilibrium transport theory for interacting quantum dots.

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[18] Neglecting the real part of the self-energy we find for the noninteracting case: $\alpha\Delta V^0/(\Gamma^L+\Gamma^R)=2\sqrt{2}/\pi^3[7\zeta(3)-2\,\mathrm{Re}\,\zeta(3,1/2+i\ln(1+\sqrt{2})/\pi)]=0.6678$ with small deviations for different polarities in the interacting case.