Optical Blocking of Electron Tunneling into a Single Self-Assembled Quantum Dot

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Time-resolved resonance fluorescence (RF) is used to analyze electron tunneling between a single self-assembled quantum dot (QD) and an electron reservoir. In equilibrium, the RF intensity reflects the average electron occupation of the QD and exhibits a gate voltage dependence that is given by the Fermi distribution in the reservoir. In the time-resolved signal, however, we find that the relaxation rate for electron tunneling is, surprisingly, independent of the occupation in the charge reservoir—in contrast to results from all-electrical transport measurements. Using a master equation approach, which includes both the electron tunneling and the optical excitation or recombination, we are able to explain the experimental data by optical blocking, which also reduces the electron tunneling rate when the QD is occupied by an exciton.

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Electron tunneling into semiconductor quantum dots (QDs) has been used to study Coulomb [1] and exchange interaction [2], as well as to prepare, read-out, and manipulate spin states [3,4]. It has also been employed to study shot noise [5,6] and reveal the Fano factor [7] in mesoscopic systems. Most of these transport measurements have been performed on semiconductor QDs, which were defined in a two-dimensional electron gas (2DEG) by lithography techniques [8]. Another QD system, which is highly interesting for optical purposes, are epitaxially grown self-assembled QDs [9], where the optical excitonic transitions can be coupled to a photon light field to study quantum optics [10], e.g., in resonance fluorescence [11,12]. They are also extensively studied for optical devices, like single photon sources [13–15], QD lasers [16,17], or optical amplifiers [18].

We use here resonance fluorescence (RF) as an optical probe to study the transport tunneling dynamics between an electron reservoir and a single self-assembled QD. Using voltage pulses and a time-resolved RF detection scheme, we are able to measure the relaxation rate for tunneling between the QD and the charge reservoir. We find clear evidence that the optical excitation of the QD reduces this rate, effectively leading to an optical blocking of single electron tunneling.

The investigated sample was grown by molecular beam epitaxy (MBE) and resembles a field-effect-transistor structure [1,19] containing a layer of self-assembled InAs QDs (see also Ref. [20]). We use a confocal microscope setup in a bath cryostat at a temperature of 4.2 K (see also Ref. [21]). RF spectroscopy of the exciton X and trion X^- resonances at different gate voltages and laser frequencies shows a transition region between 0.26 and 0.27 V (outlined in red in Fig. 1). In this region, tunneling of an electron between the QD and the charge reservoir is possible [24] and both transitions are simultaneously visible. Because of the thermally broadened distribution of electrons in the back contact, in this range of gate voltages, the QD is occupied by a single electron with a probability P, giving rise to the X^- transition. Correspondingly, the QD will be empty with a probability $1-P$ leading to the observation of the X transition.

A time-resolved RF measurement scheme is used to investigate the tunneling of a single electron into a single QD by an n-shot measurement [25]. For each shot, we first prepare an empty QD state by setting the gate voltage to $V_1 = 0$ V, well below the transition region. The laser energy is adjusted so that RF will occur for a gate voltage V_2, which lies within or near the transition region. Therefore, as long as V_1 is applied to the gate, no RF signal is observed,—see times $t < 0$ in Fig. 2(a). At $t = 0$, the gate voltage is switched to V_2, which influences the QD in two ways: (i) On the one hand, it shifts the QD excitonic transition by the quantum confined Stark effect [26] into resonance with the laser energy and we observe a RF signal by resonant light scattering (RF signal in Fig. 2 for $t \gtrsim 0$ ms); (ii) on the other hand, it shifts the energy levels

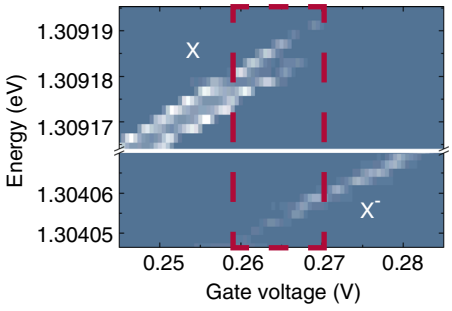

FIG. 1. Resonance fluorescence (RF) scan of the exciton (X) (with a fine structure splitting of about 8 μeV) and trion (X^-) for different laser excitation energies and gate voltages.

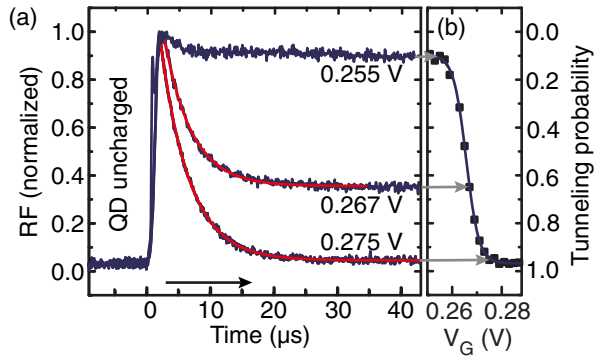

FIG. 2. (a) Measured time-resolved RF signal of the exciton transition (trion out of resonance) for three different charging voltages V_2 (red lines are exponential fits, used to obtain relaxation rates). (b) Normalized RF signal at $t = 40$ μs as a function of gate voltage.

of the QD with respect to the Fermi distribution in the reservoir. Tunneling can occur when occupied states in the electron reservoir match in energy with empty states in the QD and there will be a nonvanishing probability that the QD will be occupied by one electron. The additional electron switches the exciton transition off, as the transition for a charged QD (the X^- transition) is out of resonance with the laser energy. The evolution from an empty QD (at $t = 0$) to a thermal distribution of the QD charge at $t \to \infty$ is observed as an exponential decay in the RF signal [see Fig. 2(a)], when averaged over typically 10^6 voltage pulses.

In Fig. 2(a), we display the averaged electron tunneling signal on a microsecond time scale for three representative voltages $V_2 = 0.255$, 0.267, and 0.275 V. The time evolution of the normalized RF signal is nearly constant for $V_2 = 0.255$ V as no tunneling into the QD is possible. For a gate voltage $V_2 = 0.267$ V, an exponential decay of the RF signal is observed that saturates slightly below a value of 0.4. At this voltage, 60% of the measurements end in a situation where one additional electron has tunneled into the dot and the X emission quenches. At a gate voltage $V_2 = 0.275$ V, we observe a RF signal of the X only at the beginning of the transient, and it is completely quenched at $t = 40$ μs, indicating that the dot will be occupied by one electron with almost 100% probability at this gate voltage.

We changed the voltage V_2 in small steps (2 mV) from $V_2 = 0.252$ to $V_2 = 0.288$ V and measured the transient of the electron tunneling as discussed above. The black dots in Fig. 2(b) show the equilibrium amplitude of the RF signal at $t = 40$ μs as a function of the gate voltage. The blue solid line in Fig. 2(b) is a fit to the data with a Fermi distribution $f(E)$ where temperature, amplitude, and chemical potential were taken as free parameters [5]. The conversion from gate voltage to energy can be done by $E = ed_{\text{tunnel}}/d_{\text{dot}}V_g = e/\lambda V_g$, where λ is the so-called lever arm, given by the thickness of the tunneling barrier d_{tunnel} and the distance QD layer to gate contact d_{dot} [1,27]. This leads to the lever-arm $\lambda \approx 7$ in our sample [20].

The temperature $T = 4.2(\pm 0.2)$ K obtained from the fit is in excellent agreement with the base temperature of the helium bath cryostat.

Evaluating the exponential relaxation rates [see red lines in Fig. 2(a)], we find a constant value of $\gamma_m = 230(\pm 30)$ ms^{-1} over the entire investigated gate voltage range. This observation is quite surprising. A theoretical model developed for transport measurements [28] on similar QDs suggests that γ_m should depend on $f(E)$, as discussed in the following.

Calculations based on a master equation show that the relaxation rate is given by [28,29]

$$\gamma_m = \gamma_{\text{Out}} + \gamma_{\text{In}}, \tag{1}$$

with γ_{In} and γ_{Out} being the tunneling rates into [transition from $0 \to e^-$ in Fig. 3(b)] and out [$e^- \to 0$ in Fig. 3(b)] of the QD, respectively. They are given by

$$\gamma_{\text{In}} = d_{\text{In}}\Gamma f(E) \quad \text{and} \tag{2}$$

$$\gamma_{\text{Out}} = d_{\text{Out}}\Gamma(1 - f(E)), \tag{3}$$

where Γ is the transition rate through the tunneling barrier and d the degeneracy of the final state. In Eq. (2), $d_{\text{In}} = 2$ to account for the doubly spin degenerate empty QD state. In Eq. (3) $d_{\text{Out}} = 1$ because there is only one possible

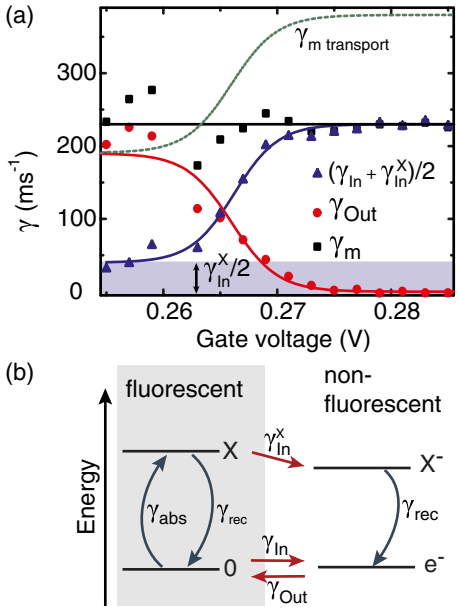

FIG. 3. (a) Bare tunneling rates γ_{In} and γ_{Out} (data points) and fits with Fermi functions (solid lines) versus gate voltage together with the measured relaxation rate γ_m (black rectangles) and the transport relaxation rate (green dashed line). (b) Energy scheme of the fluorescent and nonfluorescent states around $V_2 = 0.265$ V, where the energy of the 0 state is aligned with the energy of the e^- state. Arrows indicate optical and transport processes with their respective rates γ.

channel to discharge a singly occupied QD. Hence, $\gamma_m = \Gamma(1 + f(E))$ will be dependent on the Fermi function and therefore, on the applied gate voltage.

We explain the fact that here we do not observe an energy-dependent relaxation time with the influence of the simultaneous optical excitation. To account for the influence of the excitonic state in the QD on the tunneling rates, we extend the master equation approach [28–30] to also include the optical excitation [transition from $0 \rightarrow X$ with absorption rate γ_{abs} in Fig. 3(b)] and recombination ($X \rightarrow 0$ with rate γ_{rec}) in the QD. We, furthermore, consider the tunneling rate γ_{In}^{X} of electrons into an exciton state (transition from $X \rightarrow X^{-}$), resulting in a trion [31]. The reverse process is not possible: the energy of the trion is ≈5 meV smaller than the exciton energy (see Refs. [24,32]) so that this process would require tunneling into the back contact well below the Fermi energy, which is Pauli forbidden. Rather, we need to consider trion recombination and subsequent tunneling of the remaining single electron with rate γ_{Out} [see also arrows in Fig. 3(b)]. We distinguish between fluorescent and nonfluorescent states. The fluorescent state comprises the empty dot and the exciton state, the nonfluorescent state includes the trion as well as the singly charged QD; see left and right panel in Fig. 3(b), respectively.

To solve the Hamiltonian in first order perturbation theory, we make use of the much higher recombination rate γ_{rec} compared to the tunneling rates $\gamma_{\mathrm{In/Out}}$ (approximately 3 orders of magnitude). The time evolution of the fluorescent state is then given by the differential equation

$$\dot{P}_f(t) = -\sigma\gamma_{\mathrm{In}}P_f(t) + \gamma_{\mathrm{Out}}P_{nf}(t) - \gamma_{\mathrm{In}}^{X}(1-\sigma)P_f(t), \quad (4)$$

where P_{nf} and P_f are the occupation probabilities for the nonfluorescent and the fluorescent state and $(1-\sigma)$ is the average exciton occupation of the QD in the fluorescent state with

$$\sigma = \frac{\gamma_{\mathrm{rec}}}{\gamma_{\mathrm{abs}} + \gamma_{\mathrm{rec}}}. \quad (5)$$

By the laser excitation power, σ is tunable between 1 (weak perturbation, i.e., no exciton inside the dot) and 0.5 (saturation, i.e., the QD is occupied by an exciton half of the time).

The boundary conditions $P_f(0) = 1$ and $P_f(t) + P_{nf}(t) = 1$ are used to solve Eq. (4). We obtain

$$P_f(t) = \frac{(\gamma_m - \gamma_{\mathrm{Out}})e^{-\gamma_m t} + \gamma_{\mathrm{Out}}}{\gamma_m}, \quad (6)$$

with the relaxation rate

$$\gamma_m = \gamma_{\mathrm{Out}} + \sigma\gamma_{\mathrm{In}} + (1-\sigma)\gamma_{\mathrm{In}}^{X}. \quad (7)$$

In the experiment, the relative fluorescence amplitude is proportional to the probability that the QD is not charged. Therefore, $P_f(t)$ directly reflects the measured transients in Fig. 2(a) with a decay constant γ_m given by Eq. (7). The term proportional to γ_{In}^{X} is constant, because this tunneling takes place well below the Fermi energy, where the Fermi function equals 1. The remaining two terms in Eq. (7) are similar to the transport relaxation rate in Eq. (1), however, with an additional factor σ. Thus, the tunable factor $0.5 \leq \sigma \leq 1$ reduces the tunneling rate under illumination (for measurements see supplemental information [33]). This optical blocking can be understood from the fact that, during the time that an exciton is present in the QD, the number of tunneling paths is reduced from 2 (spin degeneracy of the electron state) to 1.

For saturation of the X transition ($\sigma = 0.5$) this optical blocking compensates the degeneracy factor of 2 in Eq. (2) and leads to a relaxation rate $\gamma_m = \Gamma(1 - f(E)) + 0.5 \times 2 \times f(E)\Gamma + 0.5 \times \gamma_{\mathrm{In}}^{X} = \Gamma + 0.5 \times \gamma_{\mathrm{In}}^{X}$ that is independent of the Fermi energy. The prediction $\gamma_m = \mathrm{const}$ is in good agreement with our experimental findings [see black data points and line in Fig. 3(a)] and shows that tunneling between the QD and the back contact can strongly be influenced by simultaneous optical excitation of the QD. For comparison, the green dashed line shows the calculated γ_m as expected in a pure transport measurement [29].

Equation (6) can be used to determine the tunneling rate out of the QD, γ_{Out}, by the time independent offset [Fig. 2(b)]. The results are plotted in Fig. 3(a) as red dots together with the tunneling rates into the QD, $0.5\gamma_{\mathrm{In}} + 0.5\gamma_{\mathrm{In}}^{X}$, which are calculated using Eq. (7) (blue triangles). As mentioned above, γ_{In}^{X} is temperature independent and γ_{in} can be fitted with a Fermi function. Using Eqs. (2) and (3), we find $\Gamma_{\mathrm{in}}^{X} = 80(\pm 20)$ and $\Gamma = 190(\pm 10)\ \mathrm{ms}^{-1}$. Thus, the transition rate into the exciton Γ_{in}^{X} is reduced by a factor of 2.4 compared to the transition rate into the empty dot Γ. This suppression of tunneling can be seen directly in the transients of Fig. 2. For $V_G = 0.255$ V, tunneling into the empty QD is energetically forbidden and the transient only reflects tunneling into the exciton state. We observe a reduction of P_f of only 10% and calculate $\Gamma_{\mathrm{in}}^{X} = 0.2\Gamma_{\mathrm{in}}$ from Eqs. (6), (7), (2), and (3). In other words, tunneling into an exciton state is reduced compared to tunneling into an empty dot.

At present this additional optical blocking mechanism ($\Gamma_{\mathrm{In}}^{X} \ll \Gamma$) is not fully understood. One possibility would be the energy shift of 5 meV between the tunneling into the exciton and tunneling into the empty dot. However, a WKB estimate only gives a change of the barrier transparency of roughly 30 percent. The transition rate through the barrier is also dependent on the extent and the orientation of the wave function in the QD, which will be different for an excitonic and empty states. A quantitative estimate of this influence is quite challenging and beyond the scope of this Letter.

In the following, we will discuss a tunneling process where the degeneracy and the factor σ do not cancel each other. Figure 4(a) shows three representative transients in a gate-voltage region where a second electron can tunnel into

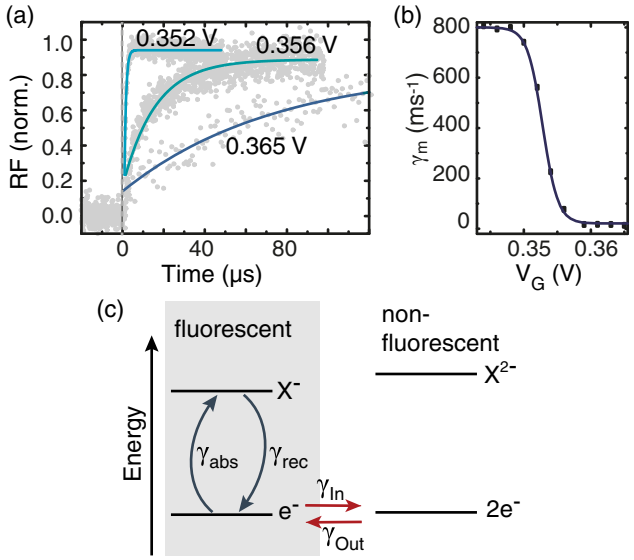

FIG. 4. (a) Time-resolved RF signal from the X^- transition, for different gate voltages when the second electron can tunnel out of the QD. (b) Plot of the experimental relaxation rates versus gate voltage with a fitted Fermi function. (c) Energy scheme of the fluorescent and nonfluorescent states around $V_2 = 0.352$ V, where the energy of the e^- state is aligned with the energy of the $2e^-$ state. Arrows indicate optical and transport processes with their respective rates γ.

and out of the QD. The measured signal is the RF of the X^- transition. We start with a gate voltage $V_1 = 0.41$ V, where the QD is charged with two electrons and, therefore, out of resonance with the laser excitation [labeled $2e^-$ in Fig. 4(c)]. At $t = 0$ we switch to a gate voltage $V_2 = 0.352$ V, where tunneling of one electron between the QD and the reservoir is possible with the rates γ_{In} and γ_{Out} for tunneling into and out of the QD, respectively. At this gate voltage, the X^- resonance matches the laser energy and we observe an increasing RF signal as the QD reaches equilibrium with the electron reservoir, and we have a nonvanishing probability of finding a single electron in the QD; see Fig. 4(a). The relaxation rates γ_m obtained from the transients [solid lines in Fig. 4(a)] are strongly gate voltage dependent. They are summarized in Fig. 4(b) and show a decrease from about 800 ms^{-1} down to almost zero. Contrary to the data shown in Fig. 3, where $\gamma_m \approx$ const, here the data can be fitted by a Fermi distribution (solid line).

To explain the gate voltage dependence of γ_m, we use again the master equation approach with the QD states: one electron, two electrons, and trion [e^-, $2e^-$, and X^-, respectively, in Fig. 4(c)]. The rates γ_{abs}, γ_{rec}, γ_{In}, and γ_{Out} correspond to the transition $e^- \rightarrow X^-$, $X^- \rightarrow e^-$, $e^- \rightarrow 2e^-$, and $2e^- \rightarrow e^-$ shown in Fig. 4(c). The solutions of the master equation give the gate voltage dependent relaxation rate

$$\gamma_m = \Gamma(2 + (\sigma - 2)f(E)), \tag{8}$$

with the degeneracies $d_{\mathrm{In}} = 1$ and $d_{\mathrm{Out}} = 2$ for the singly and the doubly charged QD, respectively. For saturated excitation ($\sigma = 0.5$), Eq. (8) suggests a drop in γ_m by a factor of 4 as the Fermi distribution in the back contact is shifted from $f(E) = 0$ to $f(E) = 1$. Experimentally, however, we find a factor of 60 [see Fig. 4(b)]. Using Eqs. (5) and (8) this leads to $\gamma_{\mathrm{abs}} \approx 18\gamma_{\mathrm{rec}}$, i.e., a strongly suppressed recombination rate. We explain this with an Auger-type recombination process [35], which results in an empty QD and leads to a suppressed RF signal until an electron has tunneled back into the QD from the reservoir. From the data in Fig. 4 an Auger rate of 20 μs^{-1} can be estimated (see Supplemental Material [36]) in reasonable agreement with the results in Ref. [35].

In conclusion, we have investigated the dynamics of electron tunneling between an electron reservoir and a single self-assembled QD under optical excitation. In contrast to transport studies, we find that the relaxation rate is independent of the chemical potential in the back contact. We explain this surprising behavior as a consequence of optical blocking, which also reduces the transition rate into the exciton state. Our findings open up a new route to optically tune the tunnel coupling between two electronic systems, a prerequisite for quantum information processing. We also note that this switching is expected to be extremely fast and only limited by the Rabi frequency of the QD exciton transition.

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