Thermal shift of the resonance between an electron gas and quantum dots: what is the origin?

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Abstract

The operation of quantum dots (QDs) at highest possible temperatures is desirable for many applications. Capacitance–voltage spectroscopy (C(V)-spectroscopy) measurements are an established instrument to analyse the electronic structure and energy levels of self-assembled QDs. We perform C(V) in the dark and C(V) under the influence of non–resonant illumination, probing exciton states up to $X^{4+}$ on InAs QDs embedded in a GaAs matrix for temperatures ranging from 2.5 to 120 K. While a small shift in the charging spectra resonance is observed for the two spin degenerate electron s-state charging voltages with increasing temperature, a huge shift is visible for the electron–hole excitonic states resonance voltages. The $s_2$-peak moves to slightly higher, the $s_1$-peak to slightly lower charging voltages. In contrast, the excitonic states are surprisingly charged at much lower voltages upon increasing temperature. We derive a rate-model allowing to attribute and value different contributions to these shifts. Resonant tunnelling, state degeneracy and hole generation rate in combination with the Fermi distribution function turn out to be of great importance for the observed effects. The differences in the shifting behaviour is connected to different equilibria schemes for the peaks–s–peaks arise when tunnelling-in- and out-rates become equal, while excitonic peaks occur, when electron tunnelling-in- and hole-generation rates are balanced.

1. Introduction

Self–assembled quantum dots (QDs) are promising candidates for future quantum technology applications as well as an interesting model system for studying fundamental quantum mechanics in a defined solid state environment. QDs can be used as suitable sources for indistinguishable single photons [1] for quantum communication [1–3], as qubits for quantum computing applications [4] or as repositories for electrons and holes, which is useful for creating new types of memory devices [5]. Also devices for energy conversion as thermoelectric energy harvesters [6] or QD solar cells [7] are envisioned. The investigation of the system’s behaviour at increasing temperature is desirable for all those applications. That requires a deeper understanding of the charge carrier transfer processes, i.e. tunnelling. Thereby, new devices operating at higher temperatures than liquid helium are in reach.

The energy level structure of QDs can be revealed by C(V)-spectroscopy [8]. Warburton et al modelled the different types of Coulomb–interaction in such systems and derived the corresponding energies theoretically [9]. Recently, it has been shown, that by creating holes in self-assembled QDs in a Schottky–diode under illumination, also excitons of different charge-state can be detected electrically [10, 11]. Bayer and Forchel [12] performed temperature dependent investigations and determined the QD’s homogenous linewidth increasing only by a tiny amount compared to $k_B T$, making QDs promising candidates for higher temperature applications. A study of the tunnelling dynamics is performed by Luyken et al [13] and a description of the differences for in- and out-tunnelling rates due to degeneracy of the various electronic states is performed by Beckel et al [14].
However, a complete report of the tunnelling dynamics of self-assembled QDs coupled to an electron reservoir and the change with increasing temperature has not been given yet. We observe and provide an explanation for a huge thermal shift of excitonic states and a smaller shift for s-peaks in the gate voltage resonance for the $C(V)$ of our QDs. We explain that shift of the peaks with electrons of energies disparate to the electrochemical potential $\mu_V$. The difference in the behaviour of the s-peaks and the excitonic peaks is interpreted as a consequence of the different resonant tunnelling conditions: a resonant tunnelling in and out equilibrium for the s-peaks and a hole generation, tunnelling-in and recombination equilibrium for the excitonic peaks.

The manuscript is organized as follows. In section 2 the sample structure and the experimental details are described. Section 3.1 deals with non-illuminated and section 3.2 with illuminated measurements. Section 4 summarizes and concludes our findings. While the physics for understanding the experiments are motivated in the paper, in the supplementary material excessive additional information on the sample structure, experimental details, the theoretical model and more experimental data can be found.

2. Sample structure and experimental details

The experiments are performed on a layer of self-assembled InAs QDs grown in the Stranski–Krastanow growth mode [15] in a molecular beam epitaxy system. The layer is embedded in a MIS-structure consisting of a 300 nm thick highly n-doped ($n = 1.8 \times 10^{18} \text{ cm}^{-3}$) back contact, an undoped tunnelling barrier of $d_0 = 25 \text{ nm}$ thickness followed by the QD-layer. A 185 nm GaAs/AlAs superlattice prevents leakage currents and increases the efficiency of hole trapping. On top of that, a 20 nm semi-transparent gold gate allows the application of a voltage as well as the illumination of the structure with an LED driven by a current $I$. The structure of the sample is sketched in figure 1 together with the corresponding band structure.

The measurements are performed in a He-closed-cycle-cryostat with a base temperature of 2.5 K. The temperature of the sample can be controlled in the range of 2.5–300 K via heating. For performing $G(V)$-spectroscopy, an AC-voltage $U_{AC} = 20 \text{ mV}_{\text{eff}}$ is added to a DC-gate-voltage, which results in the charging and discharging of the capacitor consisting of the back contact and the gate. The 90° phase shifted charge current is measured using a lock-in-amplifier. When electrons in the back contact come into resonance with the energy levels of the QD, they are able to tunnel into the dot, which leads to an increased charging current and therefore a local maximum in the $C(V)$-spectrum. The applied gate voltage can be converted into an energy using the geometrical lever arm approach [16] relating the total length between back contact and gate $d_{\text{tot}}$ to the nominal tunnel length $d_0$. In our sample this quantity is $d_{\text{tot}}/d_0 = 8.4$.

3. Results and interpretation

3.1. s-peaks

In figure 2(a), the spectra of measurements at non-illuminated (dark) conditions around the position of the s-peaks are shown. Those two charging peaks originate from electron charging of the QD ensemble with the first
and second electron on an s-shell. The peaks are separated by the Coulomb repulsion, lifting the spin degeneracy and broadened due to the ensemble inhomogeneity, i.e. the QDs are not identical and get charged at slightly different gate voltages. The peaks broaden even more with increasing temperature. A fluctuation of the QDs eigenenergies due to interaction with the phonon bath leads to a temperature dependent broadening of up to 0.1 meV at 100 K [12], certainly not enough to explain the observed effect. We attribute the main contribution to the smeared Fermi distribution in the back contact. The charging state of a QD thus underlies a certain statistical probability with an effective width of $k_B T$.

Looking more carefully to the spectra, it appears that upon temperature rise the s$_1$-peak shifts to lower voltages, while the s$_2$-peak does the opposite (solid arrows in figure 2a). To explain this, we will first exclude any significant peak-shift due to Coulomb repulsion between the electrons from electrostatic origins. Then, we apply our model.

An outward peak shift could be explained by an unpredicted increased Coulomb repulsion. Parameters that change with temperature are the dielectric constant (increase [17]), the lattice parameter (increase [18]), the effective mass (decrease [19]), the confinement energy (barely any change) and the bandgap energy (decrease [20]). In the Coulomb integral [9], the dielectric constant is in the denominator and would thus lead to a reduced Coulomb repulsion. A larger lattice parameter or/and a lower effective mass would extend the electron wave functions and the outcome is the same. The peaks would shift inwards. A change in the carrier binding to first approximation would move both peaks parallel, which is not observed. As we restrict our observation to the conduction band, a change in the bandgap would lead to a parallel shift, if the Schottky barrier height is altered. As the peaks move outwards and do not shift parallel or move inwards, we can disclaim all these hypotheses.

Our explanation of the observed effect includes a consideration of the tunnelling rates. The resonance condition, i.e. the peak in the $C(V)$-spectrum occurs, if there is an equilibrium between the respective charge states $m$ in the QDs. A simple argument is the following: the transition and thus peak called s$_1$ occurs when $m$ changes from 0 to $-1$. Thus the probability to find both charge states is identical and equal to 1/2. To fulfil this, the tunnelling rates into the QD and tunnelling rates back to the reservoir have to be equal.

$$ \Gamma_{\text{in}} = \Gamma_{\text{out}}. $$

(1)

The rates for the tunnelling processes from a charge state $\tilde{m}$ into a charge state $m$ at a certain energy are given by:

$$ \Gamma_{\tilde{m} \rightarrow m} \propto g_{\tilde{m} \rightarrow m} T_{\tilde{m} \rightarrow m}(E) f(E, T) D(E), \quad \tilde{m} > m $$

(2)

for electron tunnelling-in, and

$$ \Gamma_{m \rightarrow \tilde{m}} \propto g_{m \rightarrow \tilde{m}} T_{m \rightarrow \tilde{m}}(E) [1 - f(E, T)] D(E), \quad \tilde{m} < m $$

(3)
for electron tunnelling-out. \( g \) is the degeneracy of the respective state in the QD, \( T(E) \) is the tunnelling probability, \( f(E, T) \) is Fermi’s distribution function and \( D(E) \) is the density of states in the back contact. The first two terms on the right hand side describe the tunnelling probability for one electron. The latter two factors in the product give the density of electrons in the back contact at a certain energy \( E \) for \( G_{in} \), or the density of free states in the back contact for \( G_{out} \), respectively.

For the \( s_1 \)-state, an electron with arbitrary spin orientation can tunnel into the dot, corresponding to a twofold degeneracy \( \leftrightarrow \ (g^s_{2,1} = 2) \), while if the dot is occupied, there is either a spin-up or a spin down electron in the QD, which means \( g^s_{1,1,0} = 1 \) for tunnelling out. The peak condition (1) at a resonance energy \( E_{\text{res}}^{s_1} \) together with the in- and out-tunnelling rates (2) and (3) is solved by:

\[
f (E_{\text{res}}^{s_1}, T) = 1/3.
\]

Calculating the resonance energy peak shift for this value of the Fermi distribution yields:

\[
E_{\text{res}}^{s_1} = \mu_F = \ln (2)k_B T : = m_1 T.
\]

For the \( s_2 \)-state the QD is already filled with an electron of a certain spin orientation. According to that, only an electron with opposite spin may enter the dot and tunnelling-in is not twofold spin degenerated \( (g_{-1,0,1} = 1) \). The two electrons in the dot are now energetically degenerated and thus there are two possibilities of spin directions leaving the dot. Therefore, tunnelling-out is twofold spin degenerated \( (g_{-2,1,0} = 2) \). This leads to:

\[
f (E_{\text{res}}^{s_2}, T) = 2/3,
\]

\[
E_{\text{res}}^{s_2} = \mu_F = - \ln (2)k_B T = : m_2 T.
\]

This is an important finding, as for both states \( s_1 \) and \( s_2 \) the resonance condition is not fulfilled for the position of the Fermi energy \( \mu_F \) at \( f(E) = 1/2 \), as one could expect. Instead, the resonant points shift to higher energies for \( s_1 \) and lower energies for \( s_2 \) due to the difference in their tunnel-in and -out degeneracy. This corresponds to lower respectively higher gate voltages. It is worth to mention that the resonance peak shift is independent of material parameters and purely a consequence of the Fermi distribution and the level of degeneracy in the QDs. It resembles the entropy of a two level system.

The observed and calculated shifts are shown in figure 3(a). The slopes calculated according to the simple lever arm approach [16] are \( m_1 = 0.998(22) \times \ln (2)k_B \) for \( s_1 \) and \( m_2 = -0.919(33) \times \ln (2)k_B \) for \( s_2 \), which is in good agreement with our model. The tendency to smaller slopes for the \( s_2 \)-peak, especially at higher temperatures is qualitatively explained by the changes of the electrostatic environment changing the Coulomb repulsion (see disclaimed hypotheses above).
3.2. Excitonic-peaks

The spectra of excitonic charging peaks for different temperatures are shown in figure 2(b). The sample has been illuminated with an LED current of 2 mA and the spectra have been measured with an AC-frequency of 529 Hz. The positions of the peaks are plotted in figure 3(b).

Such an excitonic peak appears at low temperatures, when the Fermi energy is aligned to the electron eigenenergy in the QD charged by a number of holes via illumination [11]. These eigenenergies originate from attractive Coulomb interaction of the holes and the electron and thus appear at lower gate voltages than the s-charging peaks.

With increasing temperature, all excitonic charging peaks are shifted to lower voltages. The shift is huge compared to that of the s-peaks. For $X^0$ the variation is nearly 1 V of gate voltage in the range from 2.5 to 120 K, whereas both s-peaks shift for $\sim 40$ mV in this temperature range—a difference of more than one order of magnitude.

We attribute the main reason to a different peak creation mechanism. For the excitonic peaks, only intra-tunnelling of electrons is relevant. The tunnelling-out processes, which counteract the peak shifting in case of $s_1$, or even invert it for $s_2$, do usually not appear, as the recombination time of an electron hole pair is on the order of one nanosecond [3], orders of magnitudes faster than the AC period of $\sim 2$ ms in our experiment.

In contrast to the tunnelling-out process of the s-peaks, an excitonic charging peak can only occur, if a hole is generated in the QD. This generation is the equivalent of an electron out-tunnelling event. The resonance condition is fulfilled, when the electron tunnelling-in rate $\Gamma^e_{1\rightarrow 0}$ for one electron tunnelling into a single positively charged QD and the hole generation rate $\Gamma^{h, gen}_{0\rightarrow 1}$ where one hole is generated from an uncharged QD, are equal:

$$\Gamma^e_{1\rightarrow 0} = \Gamma^{h, gen}_{0\rightarrow 1}.$$  \hspace{1cm} (8)

The hole generation is a complicated process. Electron hole pairs can be created in different parts of the sample: the tunnelling barrier, the wetting layer or directly in the QDs. A portion of the generated electron hole pairs will certainly directly recombine. However, a significant amount of electrons drift, diffuse or tunnel into the back contact, while the holes relax into the QDs. A stronger illumination creates more electron hole pairs, while a more negative gate voltage makes it easier for electrons to leave the QDs. A precise expression for the rate is unknown. As we expect no strong change with applied gate bias, as an approximation we restrict our analysis to a constant hole generation rate.

The tunnelling in rate of electrons for the excitonic recombination $\Gamma^e_{1\rightarrow 0}$ might originate from the following three contributions:

1. Electrons entering the QDs above the edge of the tunnelling barrier conduction band.
2. Spatially indirect recombination of an electron in the back contact with a hole in the QD [21].
3. Resonant tunnelling in QD states.

Mechanism 1 should be negligible, because the energy to overcome is typically 250 meV and thus the number of electrons beyond the conduction band edge is low at the investigated temperatures.

The second mechanism to be considered is the spatially indirect recombination. Electrons in the back contact and holes in the QDs are quantum mechanical particles represented by wave functions. According to Fermi’s golden rule recombination can take place, if there is an overlap in the wave functions. That means electrons in the back contact can recombine with holes without having a resonant state in the QD. This recombination is usually much slower than spatially direct recombination [21]. In our case, it might play a crucial role as this time might become comparable to the inverse tunnel rate. We develop a rate equation (see supplementary) to attribute this annihilation process.

$$\Gamma^{e, indirect}_{1\rightarrow 0} \propto f(E, T) \langle \Psi_{BC}^e | c_{12} | \Psi_{QD}^h \rangle^2,$$ \hspace{1cm} (9)

where the terms of the dipole matrix element are the dipole strength $c_{12}$, the electron wavefunction in the back contact $\Psi_{BC}^e$ and the hole wavefunction in the QD $\Psi_{QD}^h$. An analysis of the gate voltage dependency yields a smooth bias dependency of the rate (see figure 3, supplemental material). Labud et al [11] show, that the excitonic peak-position is barely hole generation rate dependent by varying the illumination intensity nearly two orders of magnitude, while the smooth bias dependence would imply such a dependency. Thus this spatial indirect recombination has to be less important and another effect with a steep increase in rate must be responsible for the observed excitonic peak and its temperature shift.

Effect 3, the resonant tunnelling, only happens, when an electron is resonant to a QD’s energy level. The electron tunnels resonantly into the dot, relaxes to the ground state’s energy level within a few picoseconds [22] (if not already tunneled into the ground state in the first place) and recombines with the hole. The
recombination takes place in a time of a few nanoseconds [3], much faster than the tunnelling time of the electron in the range of a few microseconds [13]. Therefore, resonant tunnelling is the limiting process and recombination happens efficiently and fast afterwards.

The tunnel in rate for the resonant tunnelling-in hole occupied states is given by an expression formed by contributions of resonant tunnelling into s-, p-, d-, f-... states and the corresponding degeneracies, also see supplementary for a derivation and plot.

\[
\Gamma_{\hat{n}\rightarrow m}^e \propto f(E, T) D(E) \sum_{j=s, p, d,...} g_j^{\hat{n}\rightarrow m} T_j^{\hat{n}\rightarrow m}(E), \quad \hat{n} > m.
\]  

A steep increase of several orders of magnitude is found at low temperatures whenever a level comes in resonance with the electrochemical potential of the back contact. This steep increase certainly gives rise to a sharp peak in the \(C(V)\)-spectra at practically constant bias condition even for a large variation in hole generation rate. In other words: the equation \(\Gamma_{1\rightarrow 0}^e = \Gamma_{0\rightarrow 1}^{\text{gen}}\) and thus the peak condition in the \(C(V)\)-spectra is fulfilled for a wide range of hole generation rates at nearly the same bias condition. With increasing temperature, the steep slope smears out due to contributions of electrons tunnelling into the QDs at higher energies and with a much shorter tunnel barrier, explaining the shift in the peaks figures 2 (b) and 3 (b). The discrepancy of our simple model to our experimental findings at temperatures above 80 K might be a consequence of a larger hole generation rate due to a higher electron out-tunnelling rate during the generation process. Neglected before, electron tunnelling-out directly after tunnelling-in without electron–hole annihilation might happen as well, if the electron tunnelling-out rate at high reverse biases becomes the same order of magnitude as the recombination rate in the QD.

Overall, our model reproduces well the large \(\lambda^0\) peak shift of exciton annihilation observed in the temperature dependent \(C(V)\)-spectroscopy experiment.

This finding is applicable to other fields where a charge reservoir is coupled to a QD and elevated temperatures are desired, like thermoelectric energy converters, QD solar cells or electrically driven or controlled deterministic solid state single photon sources. The specifications for energy barriers needed to operate the latter at elevated temperatures will be challenging. In our experiment we used a triangular shape barrier, promoting the observed severe excitonic energy shifts, as the thermally excited carriers involved in the resonant tunnelling process are much closer to the QDs (see figure 1). For rectangular barriers, the length of the tunnel barrier would not change with applied bias and the effect is thus anticipated to be much less pronounced. Other options come along with energy filters in the barrier by a type-III broken gap band line up, quantized states and k-space overlap engineering [23].

As the observed s-state shifts relies on fundamental constants only, a metrology method for determining \(k_B\) or use as a sensitive thermometer is possible.

4. Summary and conclusions

Self-assembled QDs are investigated by \(C(V)\)-spectroscopy under illumination and various temperatures. We observe thermal shifts for s-states as well as for excitonic states. The shifts are a consequence of the change in Fermi’s distribution function and resemble the entropy of a two-level system. For the s-states, we found the differences in the degeneracies of tunnelling-in and -out of the QDs to be a crucial factor. In contrast to intuition, the resonant tunnelling condition does not occur at a value of the Fermi distribution function of \(f(E, T) = 1/2\), but at \(f(E_0^{\text{res}}, T) = 1/3\) (and \(f(E_2^{\text{res}}, T) = 2/3\)) for the \(s_1\) (and \(s_2\) state, in good agreement with our measured values. Especially, the \(s_2\)-peak shifting unexpectedly opposite to the \(s_1\) peaks could be understood. A significantly stronger shift for excitonic peaks was explained by a different mechanism. After electron tunnelling-in, tunnelling-out as a counteracting factor cannot take place due to a fast recombination process. In contrast, the hole generation rate is the counteracting process here with a completely different energy dependency from the tunnelling processes, yielding the observed strong shift. The rate equations for the peak shifts could be deduced from an overlying master equation, which gives a complete picture.

The data shows that the occupancy of the eigenstates in the QDs is also understood at temperatures exceeding liquid nitrogen, extending their application potential in various fields as energy harvesting, metrology and quantum information technologies. Also limitations and the great challenge towards electrically driven single photon sources operated at elevated temperatures are quantifiable by our findings.
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Author contributions

FB and AL performed the experiment, interpreted the data and wrote the manuscript. AL grew the heterostructure, developed the model and led the project. ADW supervised the research. All Authors contributed to the manuscript and discussed it to its final form.

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