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Voltage-controlled linewidth of excitonic transitions in a single self-assembled quantum dot

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Abstract

We report electron and hole tunnelling phenomena in a single self-assembled quantum dot as a function of the applied electric field. We use absorption spectroscopy which allows us to measure excitonic transitions under conditions where optical recombination cannot be observed due to the high, ionizing, electric field.

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Keywords: Quantum dots; Linewidth; Voltage-control; Absorption

Based on their promising applicability in quantum computation [1–3] and quantum communication [4–6], self-assembled semiconductor quantum dots (QDs) have been the focus of research for several years. Their atomic-like discrete energy states combined with their incorporation in a semiconductor heterostructure allows the study of the optical properties of single quantum dots, tuned by magnetic and electric fields, in a very controlled way [7–10]. Recently, tunnelling dynamics of electrons controlled by an electric field have been demonstrated [11]. In this paper, we report not only electron tunnelling to the continuum of the back contact but also resonant hole tunnelling phenomena to a two dimensional hole system. We show that the tunnelling mechanisms are strongly dependent on the applied electric field. Hence the lifetime limited linewidth can be controlled directly by a gate voltage.

In the experiment we present here, we investigated the excitonic transitions in self-assembled InAs/InGaAs quantum dots that are embedded in a field effect structure [12]. In Fig. 1 the band structure of the sample is shown schematically. The quantum dots were grown with molecular beam epitaxy. They are separated by a 25 nm thick GaAs tunnel barrier from the back contact (highly n-doped GaAs). The QDs are overgrown with 30 nm of GaAs. In order to prevent charge transfer from the semitransparent metallic top gate (5 nm of NiCr), a 120 nm thick blocking barrier consisting of a periodic AlAs/GaAs superlattice was deposited above the QDs. With this geometry, the electric field the QDs experience can be tuned in a controlled way, simply by applying a dc voltage between the back contact and the top gate. Furthermore, the QDs can be charged with single electrons by shifting their electronic levels beneath the Fermi energy [7,9].

In this letter we concentrate on the neutral exciton only. The measurements were carried out with Stark-shift modulation absorption spectroscopy [13,14]. This allows us not only to measure homogeneous linewidths of the excitonic transitions but also to measure at gate voltages where the QD is in the ionization regime. This is the regime where the tunnelling barrier to the back contact (which is voltage dependent) has become so small that the tunnelling rate of the electron in the QD is higher than the recombination rate. If the electron tunnels out of the QD

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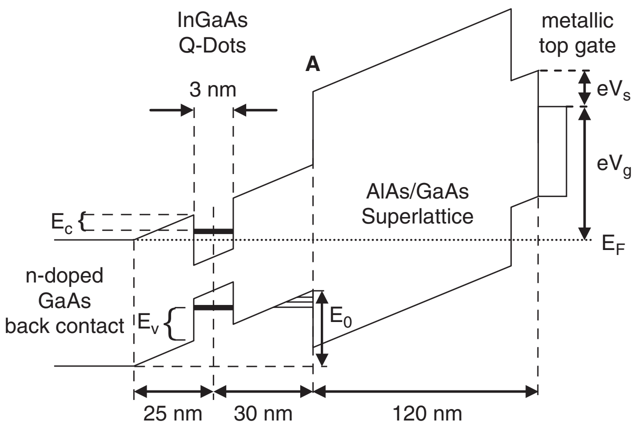

Fig. 1. Scheme of the energy band structure of the sample along the growth direction. The voltage V_g allows to control the electric field at the position of the QD. E_F is the Fermi energy, E_c and E_v are the conduction and valence ionization energies, respectively. A labels the interface between the capping layer of GaAs above the dots and the AlAs/GaAs superlattice. E_0 is the energetic point of origin of the triangular potential well created at this interface.

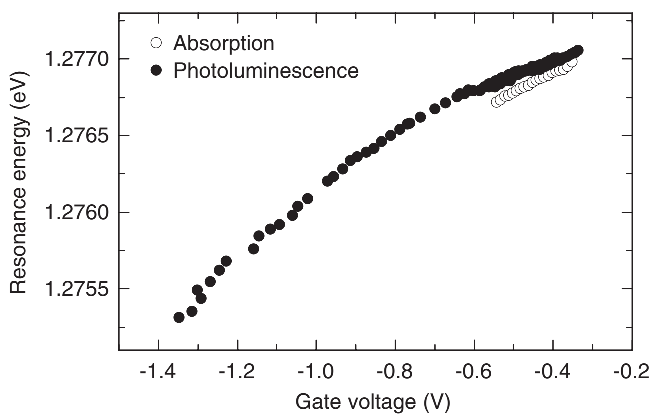

Fig. 2. Resonance energies of the neutral exciton (X^0) as a function of the applied gate voltage. The absorption data are represented by the black dots, the PL by the open circles. The small discrepancy in energy between the absorption and emission data is due to instrumental reasons.

before it can recombine with the hole under emission of photoluminescence (PL), the PL is quenched [17–19]. However in absorption spectroscopy an exciton is created under absorption of a photon. This can still happen, even if the electron (and the hole) tunnel out of the QD before they can recombine optically. The shorter lifetime of the exciton in the QD will in this case lead to a broader linewidth than in the gate voltage region where PL is observable and the linewidth should represent the lifetime limited by spontaneous emission. The experiment was performed with a confocal microscope at cryogenic temperatures.

The energy dispersion of a neutral exciton (X^0) is shown in Fig. 2. Here the black dots represent the resonance energies of the neutral QD, measured in absorption. The fine structure of the neutral exciton is of the order of 20 μeV and cannot be resolved in this graph [14]. The open dots represent the PL data of the very same dot. The voltage extent of the X^0 PL shifts to more negative voltages with increasing excitation power [15]. In Fig. 2, we plot the PL positions extrapolated to very low powers. The origin of this behavior is hole storage at the interface to the blocking barrier (A in Fig. 1). The holes, a space charge, screen the applied electric field [15,16].

It can be clearly seen that, as expected, the excitonic transition can be observed to far more negative gate voltages in absorption, than in PL. Two absorption spectra are shown in Fig. 3. At gate voltages where PL can be observed we measure very narrow linewidths as small as 1.2 μeV (Fig. 3(a)). The reason why the measured linewidths are always larger than the expected values is due to fluctuations of the resonance energy of the excitonic transitions of the order of the linewidth [14]. At large negative gate voltages, at which no PL can be observed, the linewidths are up to two orders of magnitude larger (Fig. 3(b)).

In Fig. 4 (a) the measured linewidth is shown as a function of gate voltage. We observe an exponential increase of the linewidth with decreasing gate voltage. Further oscillations of the linewidth are noticeable this will be discussed later. In the regime (ionization regime) where no PL is observed ($V_g < -0.54$ V) we conclude that the lifetime of the excitonic state is limited by the tunnelling

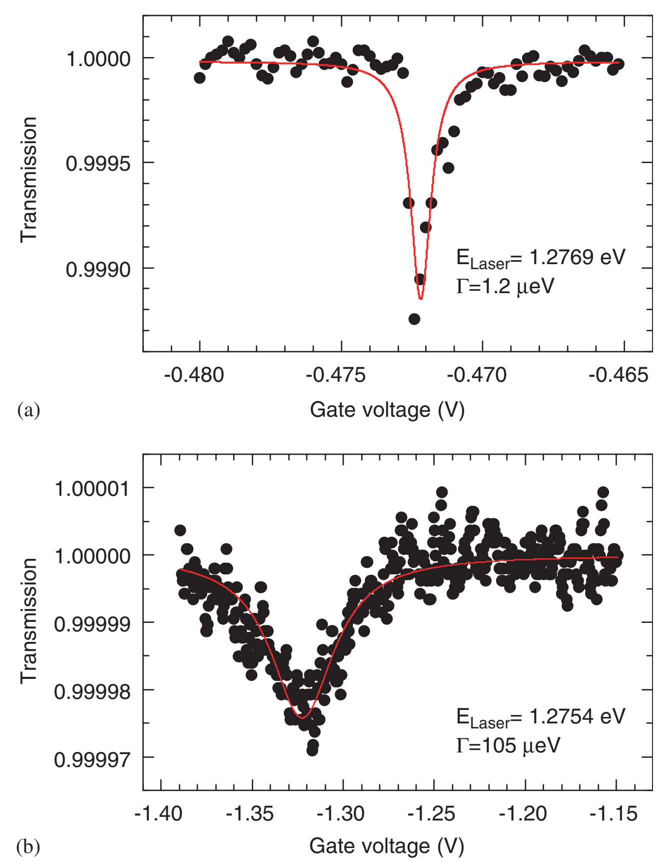

Fig. 3. Two absorption spectra measured at two different gate voltages. The linewidth can be calculated via the Stark shown in Fig. 2.

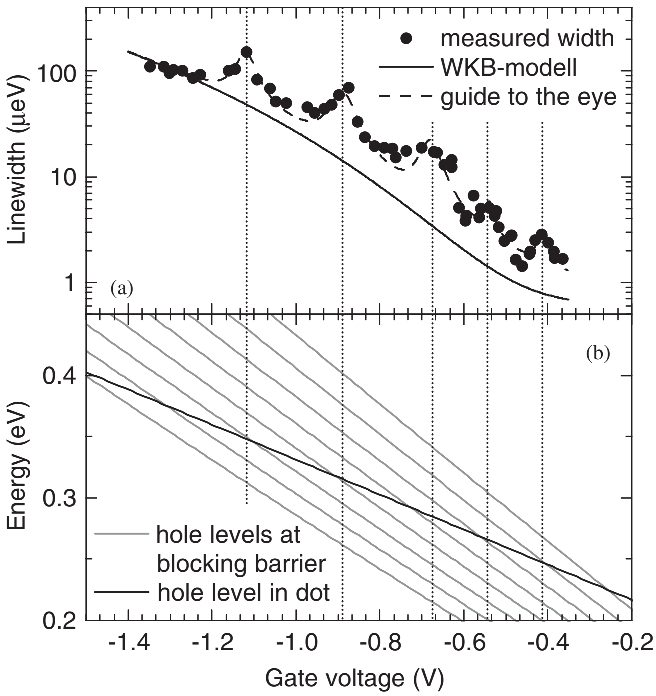

Fig. 4. (a) Shows the linewidth as a function of gate voltage. The solid line corresponds to the electron tunnelling that has been modelled with a WKB approximation, (b) represents the hole energy levels E_h in the QD (black) and E_n in the triangular potential well formed at the interface A (Fig. 1) (gray). The energies are defined relatively to the valence band edge at the back contact.

time of the electron. For gate voltages $V_g > -0.54$ V, the lifetime is rather limited by the spontaneous emission of a photon (We neglect phonon assisted dephasing, as the experiment was performed under cryogenic temperatures $T = 4.2$ K, as well as power broadening due to the low excitation powers of $\approx 1\,\mathrm{W/cm^2}$ [14]) [11]. Considering a triangular tunnelling barrier between the quantum dot and the back contact as shown in Fig. 1, we can model the tunnelling time and hence the linewidth with a WKB-approximation

$$\Gamma_{\mathrm{WKB}} = \frac{\hbar^2 \pi}{2m^* L^2} \exp\left(\frac{4}{3\hbar} d(V_g)\sqrt{2m^* E_c}\right). \tag{1}$$

Here L is the height of the dot in z-direction (3 nm), E_c is the conduction band offset (Fig. 1) and $m^* = 0.07\,m_e$ is the effective mass of an electron in GaAs [10]. Also we use the gate voltage dependent tunnelling distance:

$$d(V_g) = \frac{E_c}{e(V_g - V_s)} d \tag{2}$$

with the built in voltage $V_s = 0.62$ V [21]. Fitting formula 1 with an offset $\Gamma_0 = 0.62\,\mu\mathrm{eV}$ that corresponds to the lifetime limited linewidth we get a value for the conduction band offset of $E_c = 134$ meV. This corresponds very well with the value received from analyzing the charging behavior of the dot [20].

The oscillations of the linewidth as a function of the gate voltage represent a resonant hole tunnelling effect. The AlAs/GaAs superlattice forms a triangular potential well at the interface to the GaAs capping layer (A in Fig. 1). This leads to quantized two-dimensional states in the valence band as schematically indicated in Fig. 1. The steepness of the triangular potential well and therefore the energies of the quantized states are dependent on the gate voltage. In Fig. 4(b) the energies of the quantized states in the triangular well and the hole state in the QD are plotted as a function of gate voltage. Comparing the gate voltages of the maxima of the linewidth and the crossing points of the two energy levels (the dashed lines are a guide to the eye) reveals a striking agreement. To calculate the energies in the triangular quantum well we used the result [22]

$$E_n = E_0(V_g) - c_n \left(\frac{(eF(V_g)\hbar)^2}{2m^*}\right)^{1/3}, \tag{3}$$

with $m^* = 0.5\,m_e$ the effective mass of the heavy hole in GaAs, $c_n \approx (\frac{3}{2}\pi(n - \frac{1}{4}))^{2/3}$ and the electric field $F(V_g) = (V_g + V_s)/175$ nm. The parameter $E_0(V_g) = e(V_g + V_s)/\eta'$ (with the lever arm $\eta' = \frac{175}{55}$) describes the point of origin of the potential well as shown in Fig. 1. The hole level in the dot is described by

$$E_h = e\frac{V_g + V_s}{\eta} + E_v - E_b \tag{4}$$

here the leverarm is $\eta = \frac{175}{25}$ and the valence band offset $E_v = E_c((1/Q) - 1) = 97$ meV with $E_c = 134$ meV from before and $Q = 0.58$ from [23]. The shift of the hole level in the QD by the binding energy of the exciton ($E_b = 6.2$ meV) can be calculated in an analog way and is determined to be $E_b = 2.7$ meV [20]. The hole tunneling is fast whenever a 2D valence continuum state with in-plane wave vector less than $k_\parallel^{\max} \simeq 1/r_\parallel$ is available where $r_\parallel$ is the in-plane extent of the quantum dot wave function. However, when the only available continuum state has $k > k_\parallel^{\max}$, tunnelling is suppressed. In these experiments, $r_\parallel \approx 5$ nm, implying $k_\parallel^{\max} \approx 2 \times 10^8\,\mathrm{mm}^{-1}$, yet the energetic separation between the $k_\parallel = 0$ 2D hole states is about 20 meV, such that a hole wave vector of up to $5 \times 10^8\,\mathrm{m}^{-1}$ is required away from resonance with a $k_\parallel = 0$ state. This consideration explains the strongly oscillatory nature of the hole tunnelling in Fig. 4. We note that electron tunnelling is not suppressed by the same argument as there is no strong quantization in the z-direction in the back contact.

Another argument why hole tunnelling is expected in our sample structure, is that in the ionization regime we still observe absorption. If only the electron tunnelled and the hole remained in the dot, further absorption would be blocked. Our measurements show that this is not the case. The exponential increase in the linewidth is a compelling argument for both electron tunnelling and hole tunnelling.

In summary, we have shown an electric field dependency of the linewidth of transitions to the excitonic ground state of single self-assembled quantum dots. The linewidth can be tuned over two orders of magnitude, from linewidths limited by spontaneous emission to linewidths limited by the tunnelling times of the electron or the hole stored in the quantum dot. We modelled the electron tunnelling with a straightforward WKB approximation. Oscillations of the linewidth are deduced to be caused by resonant hole tunnelling to discrete states in a triangular potential well that is formed in our sample by an AlAs/GaAs superlattice.

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