Interband absorption on self-assembled InAs quantum dots

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Abstract

We have studied the interband excitations of an ensemble of InAs self-assembled quantum dots by detecting absorption directly in transmission experiments. The dots are embedded in a MISFET structure, allowing the dots’ electron occupation to be controlled with a gate voltage. We show how Coulomb blockade in the device’s $C - V_g$ characteristic corresponds to Pauli blocking of optical transitions in transmission. Furthermore, the second absorption peak of the dots shifts by some 20 meV and weakens when the first electron level is filled with two electrons, evidence of an exciton–electron interaction. The results also provide a direct measurement of the oscillator strength of the dots. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Spectroscopy; Self-assembled systems; Quantum dots

A quantum dot has a density of states (DOS) made up of discrete peaks and this should lead to dramatic changes of the interband optical properties compared to systems with a continuous DOS. Remarkably, self-assembled quantum dots are uniform [1] enough that this discrete DOS is preserved in the DOS of an ensemble of several million dots. This can be deduced, for instance, from the series of peaks observed in photoluminescence on dot ensembles at high pump intensity [2,3]. We report here transmission experiments on an ensemble of charge-tunable self-assembled dots.

The InAs dots, grown on GaAs, are embedded in a field-effect device, as sketched in Fig. 1. The dots are separated from a back contact (GaAs n+-layer) by a tunnel barrier and from the sample surface by a blocking barrier. By applying a voltage $V_g$ between a (semi-transparent) gate on the sample surface and the back contact, it is possible to charge the dots with electrons [4]. Fig. 1 shows the $C - V_g$ characteristic of the device. Electron tunneling into the dots gives rise to a peak in the capacitance. The doublet structure at $V_g \sim -0.75$ V corresponds to tunneling of the electrons into the first state, $|0\rangle$, the splitting arising from Coulomb blockade [5]; the feature at $\sim 0.0$ V corresponds to tunneling into the second state, $|1\rangle$, where the Coulomb blockade is not completely
resolved. By integrating the capacitance over the first peak we can estimate the dot density in our sample to be \((5 \pm 1) \times 10^{10} \text{ cm}^{-2}\), the error coming from an uncertainty in the magnitude of the background signal. Transmission experiments at wavelengths \(\sim 1 \mu\text{m}\) were carried out with a Fourier transform spectrometer. The output of the spectrometer was coupled into a 800 \(\mu\text{m}\) optical fibre which guided the light to the sample in a He dewar at 4.2 K. A Ge p–i–n diode was used as detector, mounted directly behind the sample. This arrangement enabled us to observe the very weak absorption of the dots, typically 1 part in \(10^4\). We eliminated the spectral response of the setup by taking a reference spectrum at a high positive voltage when the dots are completely occupied and the interband transitions blocked.

Fig. 2 shows a series of transmission spectra taken at various gate voltages. At large and negative \(V_g\) three absorptions are visible, corresponding to the dot \(0\rightarrow0\), \(1\rightarrow1\) and \(2\rightarrow2\) excitons. \((0\rightarrow0\) stands for a transition from the \(|0\rangle\) hole to the \(|0\rangle\) electron state, and similarly for \(1\rightarrow1\) and \(2\rightarrow2\).\) A much larger reduction in transmission at higher energy comes from absorption in the wetting layer (not shown). As the first level is occupied by increasing \(V_g\) it can be seen that the \(0\rightarrow0\) transition initially weakens and then disappears altogether. This occurs exactly at the gate voltage in the \(C-V_g\) characteristic at which we can deduce that the \(|0\rangle\) state is fully occupied. This establishes the correspondence between the transport and optics and represents a direct observation of blocking due to the Pauli exclusion principle: when the \(|0\rangle\) level is fully occupied the \(0\rightarrow0\) transition is prohibited. It is confirmed by increasing \(V_g\) further so that the \(|1\rangle\) electron state is also filled, in which case we find that the \(1\rightarrow1\) transition also weakens and then disappears.

The spectra in Fig. 2 show not only Pauli blocking in the amplitudes but also shifts in the resonance energies. We concentrate initially on \(V_g = -1.2 \text{ V}\) where all the dots are empty, and on \(V_g = -0.4 \text{ V}\) where to all intents and purposes all the dots are occupied by two electrons. It can be seen that the \(1\rightarrow1\) transition is down-shifted by \(\sim 20 \text{ meV}\) and becomes weaker on occupation of the electron \(|0\rangle\) state. The energy shift is not just a consequence of a different vertical field in
the two cases as in the regime for \( V_g \leq -1.0 \), before the onset of dot charging, the 1–1 line shows no shift whatsoever (see \( V_g = -1.6 \) and −1.2 V in Fig. 2). This red-shift and weakening imply instead an exciton–electron interaction in the dots.

We can account for the energy shift by treating the Coulomb interactions between the electrons and between the electrons and holes as perturbations to the single-particle energies. In our dots the electron quantization energy is \( \sim 50 \) meV and the Coulomb interaction between two \([0]\) electrons is \( \sim 20 \) meV, justifying the approximation. When a 1–1 exciton is excited in a doubly occupied dot there are contributions to the energy of opposite sign arising from the interaction of the electron–hole pair with the two \([0]\) electrons. There is a positive Coulomb energy due to electron repulsion between the \([1]\) electron and the two \([0]\) electrons, offset to some extent by the exchange interaction between one of the \([0]\) electrons and the \([1]\) electron. Then there is a negative Coulomb interaction between the hole and the two \([0]\) electrons. Straightforwardly, the change in the 1–1 transition energy on occupation of the \([0]\) state is \( 2E_{01}^{\text{eh}} - E_{01}^{\text{eh}} - 2E_{\text{eh}}^{01} \), where \( E_{01}^{\text{eh}} \) (\( E_{\text{eh}}^{01} \)) is the direct (exchange) Coulomb energy between a \([0]\) electron and a \([1]\) electron, and \( E_{\text{eh}}^{01} \) is the Coulomb energy between a \([0]\) electron and a \([1]\) hole. We neglect any exchange interaction between electron and hole.

The confining potentials are very close to parabolic in these dots [6] which allows these energies to be evaluated using the harmonic oscillator wave functions. To simplify the calculations we also assume that the dots are disk-like, i.e. we neglect the motion in the growth direction. The parameters needed are the single-particle energies, \( \hbar \omega_e \) and \( \hbar \omega_h \), and effective lengths, \( l_e = \sqrt{\hbar/m_e^*\omega_e} \) and \( l_h = \sqrt{\hbar/m_h^*\omega_h} \), for the electrons and holes, respectively. \( \hbar \omega_e = 49 \) meV has been determined by far-infrared measurements on the same dots [5]. These authors determined also \( m_e^* \) to be 0.08 from capacitance measurements and 0.066 from far-infrared spectroscopy; we take \( m_e^* = 0.07 \) here, implying \( l_e = 47 \) Å. To determine the hole parameters, we use these electron parameters and the neutral dot 0–0, 1–1, and 2–2 transition energies. For instance, the 0–0 energy is given by \( E_g + \hbar \omega_e + \hbar \omega_h - E_{\text{eh}}^{00} \). We calculate \( E_{\text{eh}}^{00} \) with perturbation theory to give an equation connecting \( E_g \), \( \hbar \omega_e \) and \( l_h \), and similarly for the 1–1 and 2–2 transitions. We find \( \hbar \omega_h = 25 \) meV, \( m_h^* = 0.25 \) and \( l_h = 35 \) Å. The holes are therefore more localized than the electrons. With these parameters we calculate \( E_{\text{eh}}^{00} = 29.2 \) meV, in good agreement with the energy which can be deduced from the \( C - V_g \) characteristic, 26 meV (neglecting any screening effects). The other electron energies are \( E_{01}^{00} = 21.9 \) meV, less than \( E_{\text{eh}}^{00} \) as the wave function overlap is reduced, and the exchange energy is \( E_{\text{eh}}^{01} = 7.3 \) meV. The exciton binding energies are \( E_{\text{eh}}^{00} = 33.3 \), \( E_{\text{eh}}^{01} = 27.4 \) and \( E_{\text{eh}}^{11} = 22.3 \) meV which are very similar to the results of more involved calculations [6,7]. Collecting these results together we predict a shift in the energy of the 1–1 transition of −18.3 meV on complete occupation of the \([0]\) electron state, in excellent agreement with the measured −18 meV.

The reduction in amplitude cannot be so simply accounted for because the first-order change to the matrix element vanishes. Qualitatively, the Coulomb interactions between the three electrons lead to an admixture of higher energy and therefore less localized states. This reduces the overlap integral with the localized hole. Quantitative calculations would require then accurate wave functions also for the higher energy states.

A similar analysis can also be carried out for the 0–0 and 1–1 transitions with and without single occupation of the \([0]\) state. In this case the measurements are potentially complicated as the Coulomb blockade in the \( C - V_g \) is not completely resolved implying that it is impossible to choose \( V_g \) such that all the dots are occupied with one electron. However, we estimate that at \( V_g = -0.75 \) V, at least 80% of the dots are occupied singly, \( \geq 10\% \) are doubly occupied, and \( \leq 10\% \) not at all. The data at \( V_g = -0.75 \) V are shown in Fig. 2. At this voltage, the 0–0 line is down-shifted by about 4 meV as compared to the trace for empty dots, \( V_g = -1.2 \) V. Photoluminescence experiments on similar samples also give red-shifts of this magnitude [8]. The calculation predicts a shift of \( E_{\text{eh}}^{00} - E_{\text{eh}}^{00} = -4.1 \) meV for the 0–0 transition. The 1–1 transition also moves to lower energy, by about 9 meV. If the electron excited into the \([1]\) state and the stored electron in the \([0]\) state have parallel spins, the perturbation theory predicts a shift of \( E_{\text{eh}}^{01} - E_{\text{eh}}^{01} - E_{\text{eh}}^{11} = -12.8 \) meV. If the spins are anti-parallel, the exchange interaction no longer plays a role, and the shift is −5.5 meV. In the experiment, the spins are not aligned in any particular direction and the light is
not circularly polarized, presumably implying that we measure an average of these two energies, \( \sim -9 \text{ meV} \). Thus, both the 0–0 and 1–1 shifts on single occupation of the \( |0\rangle \) state are in good agreement with the experiment. We note that the red-shift in the 0–0 energy on occupation can be interpreted as the binding energy of an \( X^- \)-exciton in a quantum dot [9].

The integrated absorption can be used to measure the oscillator strength \( f \) of the dot. For the 0–0 transition we measure an integrated absorption of \( 5 \times 10^{-6} \text{ eV} \). Ignoring any local-field effects (i.e. dot–dot interactions) we can deduce an oscillator strength per dot. The integrated absorption is given by

\[
2 \int \mathcal{R}(\sigma(E)) \, dE \quad \varepsilon_0 c(1 + \sqrt{\varepsilon_r})
\]

In a semi-classical model, the conductivity \( \sigma \) can be integrated to

\[
\int \mathcal{R}(\sigma(E)) \, dE = \frac{\hbar e^2 N f}{4m_0}.
\]

The density \( N \) is twice the dot density for the 0–0 transition, and so, taking \( \varepsilon_r = 13.1 \) and a dot density of \( 5 \times 10^6 \text{ cm}^{-2} \), we arrive at \( f = 10.9 \). We can comment on this result by evaluating \( f \) according to its definition, \( f = 2(|I| \rho |F|)^2/m_0 E_p \), where \( |I\rangle \) and \( |F\rangle \) are the initial and final states involved in the transition. By writing the total wave functions as \( |F\rangle = |0_0\rangle |S\rangle \) and \( |I\rangle = |0_0\rangle (1/\sqrt{2}) (|X\rangle + i |Y\rangle) \), where \( |S\rangle \), \( |X\rangle \) and \( |Y\rangle \) are the \( s \), \( p_x \) and \( p_y \)-like Bloch functions, respectively, we have

\[
f = \frac{|\langle 0_0 | 0_0 \rangle|^2 E_p}{2E}.
\]

\( E_p \) is the Kane energy. It is clear that the maximum oscillator strength possible is for identical electron and hole wave functions in which case \( f_{\text{max}} = E_p/2E \). Taking the GaAs value, 25.7 eV, for the Kane energy \( E_p \) we deduce \( f_{\text{max}} = 11.7 \). The oscillator strength of the dots is therefore very close to the maximum possible value, a notable example of the extreme quantization achieved in this system. The integral \( \langle 0_0 | 0_0 \rangle \) is simply evaluated to give a calculated \( f = 10.7 \), in excellent agreement with the experimental result.

Finally, we have also preliminary results in magnetic field. Fig. 3 shows the transmission spectra at 0 T and at 9 T, both for electrically neutral dots.

Within our noise level, the magnetic field has no effect on the 0–0 transition, however the 1–1 transition becomes somewhat flatter in shape. The lower part of the plot is the ratio of the two spectra which emphasizes the changes due to the magnetic field. In the single-particle picture, the 0–0 line is expected to exhibit a weak diamagnetic shift with magnetic field, and the 1–1 transition should split into two. The splitting is the cyclotron energy with the electron–hole reduced mass, 18 eV at 9 T. This is certainly consistent with the experiments. However, it is clear that either smaller line widths (i.e. a small number of dots) or much larger magnetic fields are required.

To summarize, we have detected interband transitions arising from lateral confinement in self-assembled quantum dots directly with transmission experiments. Pauli blocking of the transitions is observed as the dots are filled electrically with electrons, establishing a correlation between Coulomb blockade in vertical transport and the interband optics. For transitions which are not blocked by the Pauli exclusion principle, occupation dependent shifts reveal the presence of an exciton–electron interaction. We can account quantitatively for the shifts by treating the Coulomb interactions as perturbations to the lateral confinement. We report also a measurement of the dots’ oscillator strength.

We would like to thank Dr. A.O. Govorov for very helpful suggestions and discussions. The work in
Munich was funded by the Deutsche Forschungsgemeinschaft (SFB 348) and the work in Santa Barbara by QUEST, a National Science and Technology Center under grant DMR 20007. The collaboration was funded by an EC-US award and by a Max Planck research award. One of us (R.J.W.) would like to thank the EEC for financial support through the HCM programme.

References