Infrared transmission spectroscopy of charge carriers in self-assembled InAs quantum dots under surface electric fields

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Abstract

We present a study on the intersublevel spacings of electrons and holes in a single layer of InAs self-assembled quantum dots. We use Fourier transform infrared transmission spectroscopy via a density chopping scheme for direct experimental observation of the intersublevel spacings of electrons without any external magnetic field. Epitaxial, complementary-doped and semi-transparent electrostatic gates are grown within the ultra high vacuum conditions of molecular beam epitaxy to voltage-tune the device, while a two dimensional electron gas (2DEG) serves as a back contact. Spacings of the hole sublevels are indirectly calculated from the photoluminescence spectrum by using a simple model given by Warburton et al [1]. Additionally, we observe that the intersubband resonances of the 2DEG are enhanced due to the quantum dot layer on top of the device.

Keywords: intersublevel spacings, quantum dots, 2DEG, epitaxial gates, FTIR transmission spectroscopy

Quantum-confined zero-dimensional semiconductor quantum dots (QDs) have attracted unparalleled interest for more than a decade [2, 3]. These nanostructures have an immense potential for device applications ranging from simple electronic memories [4] to novel optoelectronics [5]. The novelty can be extended by combining a quantum well (QW) with these zero-dimensional systems. Tunneling dynamics (refilling times) of electrons from QW to QD are tunable from µs to ms by changing the barrier thickness between them [2, 6].

Electronic read-out of memory devices based on self-assembled quantum dots (SAQDs) demands an effective electrical control over the device. One of the first experiments towards this goal was performed by Sakaki et al [7], where they studied the transport properties of two dimensional electron gas (2DEG) with embedded InAs QDs. Much work has been done to study the changes in conductance, carrier concentration and mobility of the 2DEG due to successive charging of QD levels [8, 9]. Electrostatic quantization results in the formation of quasi-two-dimensional electron or hole states called subbands or sublevels. The spacing between the subbands is an important parameter defining device applications. Intersublevel spacings in InAs SAQD infrared photodetectors were determined using infrared photoconductivity measurements [10, 11]. Besides, much investigation has been carried out on the intersubband transitions of the 2DEG using grating couplers [12]. However, questions arising on the influence of QDs on the intersubband spacings of the 2DEG and vice-versa in coupled nanostructures have not been addressed. Several theoretical models [13, 14] are proposed to study the sublevel spacings. Nevertheless, due to lack of experimental agreement, determination of such energetic spacings still remains a subject of intense research.

In this paper, we address these questions and study in depth the intersublevel spacings in QDs and their influence on the 2DEG intersubband spacing by Fourier transform infrared (FTIR) transmission spectroscopy via a density...
The investigated sample is grown on a semi-insulating GaAs (\textlangle{100}\textrangle) substrate using molecular beam epitaxy (MBE). Figure 1(b) schematically shows the layer sequence of the sample. An inverted high electron mobility transistor (iHEMT) structure is fabricated, on top of which InAs SAQDs are realized by Stranski–Krastanov growth mode with a nominal thickness of 2.2 monolayers (MLs), separated from the 2DEG by a tunnel barrier of a 30 nm thick GaAs layer. The QDs are then capped by 11 nm GaAs followed by a blocking barrier, which consists of 50 periods of AlAs/GaAs (3 nm/1 nm) short-period-superlattice (SPS). An epitaxial, complementary-doped and semi-transparent electrostatic gate is grown on top of the sample within the ultra high vacuum conditions of the MBE. The carrier concentration of the epitaxial gate, obtained from Hall measurements, shows values of approximately \(6 \times 10^{18} \text{ cm}^{-3}\). There are a couple of advantages for using such gates. First, these gates have better optical transmission and do not suffer from low breakdown voltages in comparison to Schottky gates. Second, they grow lattice-matched and hence induce minimal strain on the underlying semiconductor layer. A sketch of the conduction band edge is shown in figure 1(a). The scanning transmission electron microscope (STEM) image in figure 1(c) shows a cross-sectional view of typical QDs in the QD-ensemble of our device. Two QDs labeled as QD1 and QD2 show a variation in their shape. A visible change in the morphology is observed in the first three SPS over QD2 due to strain relaxation. The two dots are separated by 44 nm, however the dot period in the structure is random. An inverted etch-mask with an array of different dimensions ranging from \(200 \times 200 \mu\text{m}^2\) to \(500 \times 500 \mu\text{m}^2\) is used to prepare gates on top of the sample. The four corners of the sample are further etched by 200 nm and Indium is diffused in an inert atmosphere of Hydrogen and Nitrogen (also known as forming gas) to contact the 2DEG layer and prevent oxidation.

Interband spacings between the sublevels of electrons and holes in the InAs SAQDs (shown in figure 2) are characterized by photoluminescence measurements at three different temperatures. A modulated laser diode, which emits at 638 nm (1.94 eV), is used to excite the sample and a liquid-nitrogen-cooled InGaAs photodiode is used as a detector.
The photo-generated electrons and holes undergo energy and momentum relaxation towards the band-gap minima, where they finally recombine and emit photons with energies given by [14]

\[ E_i = E_g - E_{ei} - E_{hh,i} - E_x, \]  

(1)

where \( i \) represents the index of the levels, \( E_g \) is the band-gap of the matrix material (GaAs), \( E_{ei} \) and \( E_{hh,i} \) are the energetic distances in the conduction and valence band measured from the respective edges of the GaAs band, and \( E_x \) is the exciton binding energy. Figure 2(d) shows the schematic of the interband transitions in the QD. At 300 K, four interband transitions in QDs are observed (figure 2(a)), marked as \( E_0 \), \( E_1 \), \( E_2 \), and \( E_3 \) along with the transitions in the wetting layer (\( E_{WL} \)) and GaAs (\( E_{GaAs} \)). With decreasing the temperature to 77 K, the respective transitions shift to higher energies (figure 2(b)) according to Varshni's empirical relation [15]. Further decreasing the temperature to 10 K, only two distinct interband transitions are visible (figure 2(c)), marked as \( E_0 \) (1.062 eV) and \( E_1 \) (1.118 eV), together with the transitions in the wetting layer and GaAs. Values of QD interband transitions at three different temperatures along with their full width at half maximum (FWHM) are given in table 1.

We observe an increase in the non-normalized peak intensity of \( E_0 \) (not shown) by 50% from 300 K to 10 K. While the intensity of \( E_1 \) does not change during cooling, \( E_2 \) and \( E_3 \) disappear. On the other hand, a strong emission from the wetting layer is observed at 10 K as compared to the emission at 300 K (clearly seen from the normalized spectra in figure 2). The disappearance of \( E_2 \) and \( E_3 \) and the increase of emission from the wetting layer can be explained as follows: At low temperatures, the charge carriers in the QDs thermalize to the ground state. Also, the carriers in the wetting layer are thermalized quickly. This reduces the capture of the carriers from the wetting layer because the barrier around the QDs cannot be overcome due to low kinetic energies of the carriers. As a consequence, the number of excited state transitions is also reduced, as seen at 10 K. Moreover, the broadening of the peaks can also be assisted to the fact that Stranski–Krastanov

![Figure 2](image_url)

**Figure 2.** (a) Room temperature, 300 K, (b) 77 K and (c) 10 K photoluminescence spectra measured with an excitation power of 5 mW. Spectral deconvolutions are plotted in green. The experimental data points are plotted in grey dots and the reconstructed spectra are shown in red. (d) Schematic representation of the transitions corresponding to each peak.

| Peaks | 300 K | 77 K | 10 K |
|-------|-------|------|------|
| \( E_0 \) | 1.001 (0.036) | 1.059 (0.036) | 1.062 (0.036) |
| \( E_1 \) | 1.053 (0.038) | 1.114 (0.038) | 1.118 (0.038) |
| \( E_2 \) | 1.102 (0.047) | 1.165 (0.027) | -- |
| \( E_3 \) | 1.151 (0.066) | -- | -- |
| \( E_{WL} \) | 1.355 (0.035) | 1.422 (0.018) | 1.425 (0.013) |
| \( E_{GaAs} \) | 1.435 (0.033) | 1.512 (0.009) | 1.516 (0.006) |
corresponding to the filling of electrons in the QD levels. The red voltage. The green gaussian curves indicate the deconvoluted peaks Successive charging of QD levels is observed on increasing the gate 4.2 K. The capacitance is corrected by subtracting the linear slope. respect to the Fermi level for two gate voltages.

emission of QDs of different sizes [16].
growth of QDs leads to an ensemble resulting in a sum of the

At an intermediate voltage of 0.3 V, we see the occupation of

for the DC voltage. In the inset of figure 3, schematics of

level with an electron is observed as a change in the capacitance QDs through the GaAs tunnel barrier. The charging of the QD

as a reference voltage (V)

A

m2) is the area of the gate. The

where

is the vacuum impedance and

To rule out the long-term experimental drifts [18]. The

Direct investigation of the quantized energy levels within the conduction band is performed by infrared transmission measurements with a rapid scan Bruker IFS113V interferometer, which has two sources (mercury-arc lamp and globar) to cover a large range of infrared radiation. The spectral resolution of the measurements is 0.25 cm\(^{-1}\) (=0.03 meV). The bottom surface of the sample is wedged at around 3\(^\circ\) to avoid the Fabry–Perot interference fringes and mounted at an angle of 30\(^\circ\) (obeying the polarization selection rule for observing the intersubband transitions) in a self-built cryogenic optical sample holder, which is equipped with a liquid-Helium cooled Si-bolometer for detection. Spectra at a certain gate voltage and reference voltage are collected alternatively and averaged over long measurement times (3 h) to rule out the long-term experimental drifts [18]. The normalized transmission, \(T(V_g)/T(V_i)\), is given by [19]:

\[
\frac{T(V_g)}{T(V_i)} = 1 - \frac{2\text{Re} [\sigma(\omega)]}{(1 + \sqrt{\varepsilon + r_e/r_g}) \varepsilon_0 c^2}.
\]

where \(\varepsilon_0\) is the permittivity of free space, \(c\) is the velocity of light, \(\varepsilon\) is the dielectric constant of InAs, \(r_g = 377 \Omega\) is the vacuum impedance and \(r_e = 12 \Omega\) is the combined impedance of the epilayer gate and the back-contact at 4.2 K. The high-frequency conductivity, \(\sigma(\omega)\), of electrons in a parabolic potential is

\[
\sigma(\omega) = \frac{Ne^2 \tau}{2m^* a^2 \left[1 + \frac{(\omega^2 - \omega_0^2)}{\omega^2} \right]^2}.
\]

where \(\omega_0\) is the high-frequency resonance, \(a\) is the average

Table 2. Energies corresponding to the individual electron charging peaks in the C-V spectrum. The voltages are taken from the deconvolution result of the complete spectrum. The values in the brackets represent the energy difference between the given peak and the consecutive peak before.

| Charging peaks | Gate voltage (V) | Energy (lever arm) (meV) |
|----------------|------------------|-------------------------|
| \(s_1\)        | −1.156           | 223.1                   |
| \(s_2\)        | −0.948           | 200.5 (22.6)            |
| \(p_1\)        | −0.480           | 149.8 (50.7)            |
| \(p_2\)        | −0.327           | 133.2 (16.6)            |
| \(p_3\)        | −0.141           | 112.9 (20.3)            |
| \(p_4\)        | 0.019            | 95.5 (17.4)             |
to the experimental data. These are shown as green curves in figure 4. At \( V_g = -1.167 \) V, we observe only a small dip in the transmission due to the AlAs TO-phonon at 45.7 meV [20]. At \( V_g = -0.55 \) V, electrons from the s-sublevel absorb the incident infrared radiation and occupy the next sublevel with a minimum in the transmission spectra at 45.0 meV (\( s \rightarrow p \)), as compared to 45.1 meV calculated from the C-V measurements. Upon increasing the voltage to 0.3 V, when both s- and p-sublevels are occupied, transitions from both the levels to the d-sublevel are registered as minima in the transmission spectrum at 45.1 meV (\( p \rightarrow d \)) and 85.1 meV (\( s \rightarrow d \)), respectively. For \( s \rightarrow d \), the value is almost twice the value observed in \( s \rightarrow p \) or \( p \rightarrow d \). This is expected to a very good approximation for confinement due to parabolic potential leading to equidistant levels. However, the small variation is due to the fact that at 0.3 V the p-sublevel is also filled, leading to additional interactions, which reduce the energy. We also observe a large coupling of \( s \rightarrow p \) and \( p \rightarrow d \) transitions to the AlAs TO-phonon (45.7 meV). The coupling effect can be seen in figure 4(a) as an increase in the transmission depth of the phonon transition at 45.7 meV when a bias of \(-0.55 \) V or 0.3 V is applied as compared to the case when no transitions in QDs are observed at \(-1.167 \) V. This is an interesting feature resulting from the close vicinity of AlAs in the SPS to the QDs, separated by less than 5 nm. Using the results from PL spectra and the simple model given by Warburton et al [1], we calculate the sublevel spacing of the holes in the valence band. The energetic distance between \( h_1 \) and \( h_2 \) (see figure 2(d)) is found to be 10.9 meV. However, it is to be noted that our device structure is more complicated as compared to the one considered for developing the simple model in [1], resulting in additional complex electrostatics. The calculated hole spacing shows very close correspondence to the observed experimental value performed by Jang et al [21] via excitation-power dependence of PL spectrum for an n-modulation-doped QD structure. On the contrary, this value is small compared to the values in [22, 23], where they state that the hole sublevel spacing is in the order of 20–25 meV. Clearly, this is a riddle, which can be solved by direct access to hole sublevels via FTIR measurements on p-type samples.

At lower frequencies, strong intersubband resonances (ISRs) are observed in the 2DEG. The 2D electron density obtained from Hall measurements is \( 3 \times 10^{11} \) cm\(^{-2} \). Sufficient negative bias \(( V_r = -3 \) V) is applied to completely deplete the 2DEG below the threshold voltage, which is used as reference for the chopping scheme. The strong enhancement of the 2DEG-ISRs is due to the layer of QDs on top, which helps in better coupling of the incident radiation with the intersubband transitions of the 2DEG. This is due to the fact that the InAs SAQDs in the GaAs matrix act analogous to a grating coupler, which tilts locally the electric field vector of the incident wave, leading to substantial components in the growth direction (normal to the 2DEG), which is necessary to excite ISR. As a consequence, we observed around 6% transmission dip in the primary ISR as compared to the strengths of the transitions reported previously 1–2% [12]. With an increase in the gate voltage, the ISR shifts to higher energies owing to an increase in the carrier density steepening up the asymmetric triangular potential, which confines the 2DEG and thus leading to higher intersubband spacings.

The picture is however not that simple, since many-electron interactions significantly change the single electron intersubband transition energies of the 2DEG in a HEMT structure. Under the local density approximation and linear response theory, it was shown by Ando [24] that the two most significant contributions are the depolarization effect which blue-shifts the ISR and the exciton-like effect which red-shifts
the ISR. Besides, with an increase in the voltage ($-0.55$ V), the QDs are also charged and this will result in an additional field across the 2DEG layer. The resonances observed are a result of the combined effect of the above mentioned phenomena.

An interesting feature observed in the electronic sublevel resonances in the SAQDs via FTIR measurements is their linewidths which are as narrow as $2-3$ meV in comparison to the linewidths seen in the C-V spectrum. We observed considerable fluctuations of the thickness of the QDs in the growth direction from the STEM and TEM images (not shown). This significantly affects the ground state energy, which broadens the charging spectrum in the C-V measurements. However, the influence on the energetic spacings of $s$-, $p$- and $d$-sublevels is very small, leading to narrow linewidths in the FTIR spectra. The stability in the confining potential allows us to observe the discrete sublevel resonances in the transmission spectra of FTIR measurements, even though our experiments are performed over an ensemble of QDs [11]. Apparently, this unique property of the SAQDs has been perfected and employed over the years for device applications like QD-lasers, QD-infrared photodetectors etc.

In conclusion, the use of epitaxial, complementary doped and semi-transparent electrostatic gates to chop the density of charge carriers proved to be an efficient technique to study the intersublevel spacings of electrons in the conduction band. Using these results along with the interband transition values from PL, we evaluated the sublevel spacing of the $s$- and $p$-sublevels is very small, leading to narrow linewidths in the FTIR spectra. The stability in the confining potential allows us to observe the discrete sublevel resonances in the transmission spectra of FTIR measurements, even though our experiments are performed over an ensemble of QDs [11]. Apparently, this unique property of the SAQDs has been perfected and employed over the years for device applications like QD-lasers, QD-infrared photodetectors etc.

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