LETTER TO THE EDITOR

Size determination of InAs quantum dots using magneto-tunnelling experiments

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Abstract. Tunnelling experiments through GaAs–AlAs–GaAs structures with InAs embedded in the AlAs barrier show steps in the current–voltage characteristics which we assign to single-electron tunnelling through self-assembled InAs quantum dots between two three-dimensional electrodes. From the magnetic field dependence of the onset of the current steps, we determine the lateral extension of the electronic wave function in the dot to 4 nm, corresponding to a dot of 14 nm in diameter. Replica of steps at higher voltages are attributed to tunnelling through charged dots. A similar structural dot size is measured independently by transmission electron microscopy on the same wafer and by atomic force microscopy on control samples with InAs dots on a GaAs or an AlAs surface, respectively.

The epitaxial growth of lattice mismatched InAs on GaAs or AlAs opens new possibilities for the simple fabrication of semiconductor nanostructures. InAs quantum dots (QDs) are formed *in situ* during growth due to the relaxation of a strained InAs wetting layer on GaAs or AlAs [1]. The particular interest lies in their uniformity and small size, 10–20 nm lateral dimension and 3–4 nm height.

Previous experiments on such InAs QDs mainly concentrated on optical methods including the sharp luminescence of single dots [2]. Some experiments investigating the energy level structure of an ensemble of InAs QDs were performed using far-infrared spectroscopy [3] and capacitance spectroscopy [4].

The quantized states of a single InAs QD can be accessed in an elegant way using resonant tunnelling experiments with InAs dots embedded in a single-barrier tunnelling diode. Recently, the first of such experiments were reported [5-7]. Peaks in the current–voltage (I-V) characteristics of the diode assigned to single-electron tunnelling through individual InAs QDs were observed. The shape of the peaks is mainly determined by the energy dependent electronic density of states in the emitter.

In this letter we report on resonant tunnelling experiments through symmetric diodes with embedded InAs QDs. In contrast to all previous work [5–7] we observe clear current *steps* in *both* voltage directions of the I-V characteristics, consistent with single-electron tunnelling

between three-dimensional electrodes through a nanoscale island†.

Our QD samples were prepared by molecular beam epitaxy; first growing a strongly Si-doped, 1 μ m thick, n^+ -GaAs buffer layer on top of a n^+ -GaAs wafer $(n^+ = 2 \times 10^{18} \text{ cm}^{-3})$ serving as the back contact. The following layers are 10 nm n-GaAs ($n = 10^{17} \text{ cm}^{-3}$), 10 nm n^- -doped GaAs ($n^- = 10^{16} \text{ cm}^{-3}$) and a 15 nm undoped GaAs buffer. Between a 5 nm AlAs bottom barrier and a 5 nm AlAs top barrier 1.8 monolayers InAs were grown, which form self-assembled QDs on a wetting layer of InAs. The structure was completed symmetrically to the QDs with a GaAs buffer and a 1 μ m top contact. The strong doping in the vicinity of both barriers ensures the existence of the three-dimensional electrodes. The substrate temperature was 600 °C, except 520 °C during InAs growth. To characterize the tunnelling properties of our samples, we patterned vertical diodes of macroscopic size (10–200 μ m). The diodes were then contacted with annealed AuGeNi contacts, the n^+ -doped substrate was used as a common back contact for all diodes.

In order to clarify the structural properties of the QD samples we used transmission electron microscopy (TEM). Figure 1(a) verifies the presence of InAs dots embedded in the AlAs barrier. The dots are about 10–15 nm in diameter and their height is roughly 4 nm. We have also grown

[†] Small steps of a few pico amperes have also been observed at very low temperatures in [7] in a single bias direction.

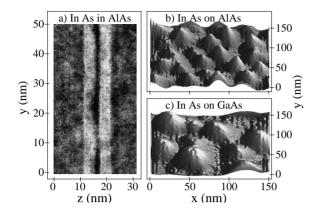


Figure 1. TEM image of InAs dots embedded in an AlAs barrier (a) compared to structural properties of InAs grown on AlAs (b) and GaAs (c) measured by AFM at room temperature.

control samples with uncovered InAs on AlAs and on GaAs under similar conditions as the embedded QD samples. For the AlAs control samples the growth was terminated after depositing the InAs on the 5 nm thick AlAs. For the GaAs control samples no AlAs barriers were grown, the InAs was directly deposited on the 15 nm GaAs buffer of the bottom contact and the growth was subsequently terminated.

Atomic force microscopy (AFM) images of such samples are shown in figures 1(b) and 1(c). Grown on GaAs and on AlAs the height of the dots is approximately 4 nm. Their lateral size is 20–30 nm for InAs dots grown on GaAs, but only 10–15 nm for the dots grown on AlAs, which is consistent with TEM measurements of structures with embedded dots. As a consequence, the density of the dots on AlAs is, with $n \approx 1000~\mu\text{m}^{-2}$, considerably higher compared to dots with an identical InAs coverage on GaAs. We attribute this fact to a higher surface roughness and subsequently shorter diffusion length on AlAs compared to GaAs. This leads to faster nucleation of InAs. Therefore, depositing InAs on AlAs instead of GaAs allows the formation and investigation of even smaller dots.

The electronic properties of the dot samples were investigated in comparison with two different reference samples. One type contains no InAs, i.e. consists of a single 5 nm/5 nm AlAs barrier with a growth interruption (of the duration that the InAs growth in the QD samples takes). In the second type a sub-monolayer of InAs is embedded in the barrier, acting as a wetting layer.

In figure 2 the I-V characteristics of a typical dot sample compared to the reference samples at liquid helium temperatures is shown. The single-barrier sample contains a dominant feature at source-drain voltages $U_1 \approx \pm 0.2$ V. We assign the increased current around these voltages to tunnelling from the Γ -band of the emitter through the state E_1^X in the X-band well of the AlAs barrier into the Γ -band of the collector, as shown in the inset of figure 2. Similar structures have been observed previously in the I-V characteristics of a single-barrier tunnelling diode [8]. We observe an additional feature at $U_2 \approx \pm 0.4$ $V = 2U_1$. We relate this to tunnelling through the X-band state E_2^X at the interface in the middle of the barrier, see inset of figure 2. Due to the growth interruption this interface is even formed in samples with no InAs. Finally, only in samples with InAs (QDs and wetting layer) an

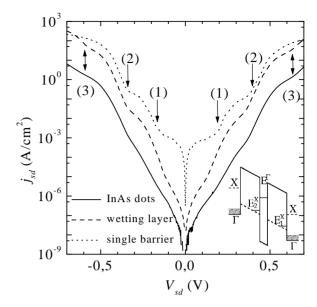


Figure 2. Current density as a function of voltage for three different resonant tunnelling diodes containing only a single AlAs barrier, an InAs wetting layer or InAs dots embedded in the AlAs barrier. The curves are shifted for clarity. The inset shows the schematic band structure of the wetting layer sample with an applied bias.

additional structure appears at $U_3 \approx \pm 0.6\,\mathrm{eV}$, it is assigned to the tunnelling through the Γ -band E^Γ of a two-dimensional InAs layer in the middle of the barrier [5].

The most interesting features appear in samples with InAs QDs in the barrier. At low voltages, typically around 100 mV, steps in the I-V characteristics appear which confirm the tunnelling process from a three-dimensional emitter to a zero-dimensional QD. An illustrative example for high temperatures is shown in figure 3. Two clear current steps are visible at voltages $V_1 = 74$ mV and $V_2 = 128$ mV. The structures are visualized more clearly in plotting the differential conduction dI/dV as a function of the applied bias voltage in the same figure. The current steps, i.e. peaks in dI/dV, persist up to a temperature of about 60 K, indicating the small size of the dot the step is due to. Two weaker current steps at 86 mV and 140 mV probably arise from dots of very similar electronic properties.

At lower temperatures (down to 300 mK), very sharp steps with a width which is mainly limited by thermal broadening are measured, indicating that the steps are in fact due to tunnelling through a single dot [9].

Steps in the I-V characteristic were observed in most of the 30 diodes of the QD samples investigated at present. However reference samples containing no dots, but an InAs wetting layer, do not show any current steps. Due to the nominally symmetric growth of the QD structures and the three-dimensional emitter we observe steps in both current directions. They appear at bias voltages of $50-200 \, \text{mV}$ with step heights between $100 \, \text{pA}$ up to a few nanoamps. We assign them to single-electron tunnelling through *individual* quantum dots. Different reasons for the selection process of an individual dot accessible to tunnelling from typically 10^5 dots present in a mesa have been proposed [5-7]. However, in our view, its precise origin still remains unclear.

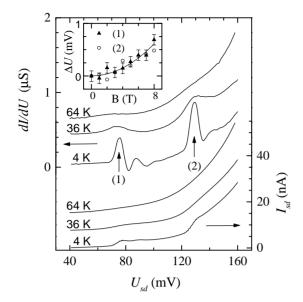


Figure 3. Fine I-V characteristics at high temperatures with clear current steps assigned to the single electron tunnelling through InAs QDs (bottom three curves). The steps are resolved more clearly as peaks in the differential conduction (top three curves). In the inset, the magnetic field dependence of the positions of the two dominant current steps (1) and (2) is shown. The full curve is a parabolic fit.

To characterize the lateral size of the dots accessed by the resonant tunnelling experiments we have performed experiments in magnetic fields $B \parallel I$. Supposing that the electrons are tunnelling through a state with quantization energy E_0 at zero magnetic field, we can deduce the lateral size of the dots from the magnetic field dependence of the position of the current steps. For low enough magnetic fields where $\hbar\omega_c\ll E_0$ ($\hbar\omega_c=\hbar eB/m^*$ is the cyclotron energy of electrons with an effective mass m^*) the change of E_0 with magnetic field is given by $\Delta E = (eBr_0)^2/(8m)$, where $r_0 =$ $\sqrt{\langle r^2 \rangle}$ defines the spatial extent of the electron wave function perpendicular to the magnetic field. This energy shift is related to a magnetic field induced shift $\Delta U_{sd} = \Delta E/(e\alpha)$ of the step position in source-drain voltage, where α is the proportion of the total voltage drop between the emitter and the dot considered. For strongly doped samples, where essentially all the voltage drops over the roughly symmetric barriers, it is reasonable to assume $\alpha \approx 0.5$.

From the diamagnetic shift shown in figure 3 the extension of the electronic wave function for the dots assigned to the two dominant steps is found to be $r_0=3.7$ nm, assuming an effective mass $m^*=0.06m_0$ (m_0 is the free electron mass) as deduced from capacitance spectroscopy [4]. Modelling the dot by a circular potential well with diameter d we deduce from r_0 a diameter d=14 nm for this particular InAs dot. Here we have already taken into account the finite height of the AlAs barrier reducing the value of d by 10% compared to an infinitely deep potential well. This electronic size is in very good agreement with the typical structural size of the dots determined from AFM or TEM†. Both steps show

the same diamagnetic shift, indicating that they are related to two dots of similar size or even the same dot.

Similar diamagnetic shifts are also observed in photoluminescence (PL) experiments on InAs dots. However, most PL experiments are performed on a large ensemble of dots, see for example [10], only few experiments exist on the diamagnetic shift in the PL of a single dot [11]. Additionally, PL probes the excitonic states, influenced by electrons, holes and excitonic interaction effects, whereas our transport experiment directly accesses the electronic energy levels of the dot. Therefore, this complementary technique is extremely useful for the understanding of the electronic structure of a single InAs QD.

For dots of the above size (14 nm), the first excited states are situated $\Delta E \approx 100$ meV above the ground state. However, due to the limited voltage range (typically V < 200 mV) where the background current is small enough to resolve steps originating from tunnelling through individual dots, we do not observe any features related to excited states of InAs dots.

Finally, it may be interesting to remark that the voltage difference between the two dominant current steps $\Delta V = V_2 - V_1 = 54$ mV coincides strongly with the charging energy $E_C \approx 28$ meV of a circular dot with $r_0 = 3.7$ nm, namely $E_C = \alpha e \Delta V$. We may therefore speculate that the two dominant current steps are related to tunnelling through the ground state of the same dot. For $V > V_1$ one electron can tunnel through the ground state of the dot, for $V > V_2$ double occupancy is energetically possible, we observe another increase in the current due to the possibility for a second electron to tunnel through the same dot.

In conclusion we have evaluated the diameter d of an individual InAs quantum dot from the diamagnetic shift of current steps due to single electron tunnelling through an individual dot. We find d=14 nm in agreement with the structural size of dots on control samples measured by AFM and TEM. The observation of a second current step can be related to tunnelling through the charged ground state of the same dot, the energy difference reflecting the charging energy of the dot.

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References

- Leonard D et al 1993 Appl. Phys. Lett. 63 3202
 Moison J M et al 1994 Appl. Phys. Lett. 64 196
- Marzin J-Y et al 1994 Phys. Rev. Lett. 73 716
 Leon R et al 1995 Science 27 1966
 Grundmann M et al 1995 Phys. Rev. Lett. 74 4043
- [3] Fricke M et al 1996 Europhys. Lett. 36 197
- [4] Miller B T et al 1997 Phys. Rev. B 56 6764 Drexler H et al 1994 Phys. Rev. Lett. 73 2252
- [5] Suzuki T et al 1997 Japan. J. Appl. Phys. 36 1917
- [6] Narihiro M et al 1997 Appl. Phys. Lett. **70** 105
- [7] Itskevich I E et al 1996 Phys. Rev. B 54 16 401
 Thornton A S G et al 1998 Appl. Phys. Lett. 73 354
- [8] Mendez E E et al 1987 Appl. Phys. Lett. 50 1263
- [9] Deshpande M R et al 1996 Phys. Rev. Lett. 76 1328.
- [10] Wilson L R et al 1998 Phys. Rev. B 57 R2073
- [11] Toda Y et al 1998 Appl. Phys. Lett. 73 517

 $[\]dagger$ As a comparison we also calculated the dot size using an effective mass $m^* = 0.023 m_0$ for bulk InAs. From this we find a diameter of the quantum dot d=8 nm which is unreasonably smaller than the structural size of the dot measured with TEM and AFM.