Magneto-photoluminescence of stacked self-assembled
InAs/GaAs quantum dots

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Abstract

We studied ten-fold stacked layers of self-assembled InAs/GaAs quantum dots by photoluminescence in pulsed magnetic fields. When the interlayer distance is reduced from 9.8 to 5.5 nm, a doubling of the diamagnetic shift for a magnetic field perpendicular to the [0 1 1] direction reveals the onset of electron coupling between the dots in the stack. On reducing the interlayer distance to 3.1 nm, a lower exciton effective mass is seen in addition to the coupling. For such a close stacking, the strain field in and around the dot is completely different from that of a single-layered structure. In particular, the strain inside the InAs dots in the stack is partially relaxed, causing the observed effect.

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1. Introduction

The self-assembly process based on strain relaxation of lattice-mismatched semiconductors can result in a wide variety of structures, such as quantum dots, wires and rings depending on the compounds used and the growth conditions [1–3]. Growing multiple layers of such structures on top of each other can increase the effective dimensionality if electronic coupling between the layers is present, e.g. closely stacking a large number of quantum dot layers produces a one-dimensional array of coupled quantum dots, i.e. a kind of quantum wire for which certain parameters can be controlled more easily than for more conventional quantum wire structures. Amongst those parameters are photoluminescence (PL) energy [4,5], radiative lifetime [4] and exciton effective mass [6].

When the distance, \( d \), between different layers in the stack is very large, each layer acts independently from the others, resulting in properties similar to those of a single-layered sample. By reducing the interlayer distance, these properties start to change due to electronic coupling between the dots, to a different dot morphology or to an altered strain profile. The coupling is studied by PL in pulsed magnetic fields. This technique has already proven its benefit in the study of stacked...
InP/GaInP [7] and single-layered InAs/GaAs systems [8]. Analysis of the diamagnetic shift of the PL energy between 0 and 50 T can provide information on the excitons confined in the stack, such as their extent and their mass [7]. We will show that in stacked InAs/GaAs dots with 1.8 monolayers (ML) of InAs in each layer, electron coupling is significant for interlayer distances ≤5.5 nm, while a significant decrease in observed mass is caused by a partial strain relaxation at \( d = 3.1 \) nm.

2. Experimental details

The stacked InAs/GaAs heterostructures were grown by molecular beam epitaxy at 450°C. After deposition of 1.8 ML of InAs on the GaAs substrate, the dots were capped by GaAs to form 9.8, 5.5 and 3.1 nm thick capping layers as measured by transmission electron microscopy for samples B, C and D, respectively. This procedure was then repeated to obtain a tenfold stack of dots. The reference sample A consists of only one such layer and was grown at 480°C.

PL measurements were performed in a helium bath-cryostat at 4.2 K in pulsed magnetic fields up to 50 T. A frequency doubled Nd:yttrium–aluminium–garnet laser operating at 532 nm focused into a 200 μm core optical fibre excited the sample with a power density of \( \sim 1 \) W cm\(^{-2}\). The luminescence collected by a bundle of six collection fibres was analysed by a 0.30 m spectrometer with a liquid-nitrogen cooled InGaAs diode array. The photon integration time was kept constant at 0.5 ms, which corresponds to a field resolution of ±0.1% at the peak of the magnetic pulse and less than ±3% during up and down sweeps.

3. Results and discussion

When the interlayer distance is reduced, a red-shift and a broadening of the zero-field PL are observed (Fig. 1). In the literature, both red- and blue-shifts have been reported for similar structures [4,5], where a red-shift can be caused by a relaxation of the strain in the dot [7,9], electronic coupling [10,11], or just by the formation of larger dots due to the stacking [12]. The line width is, in contradiction to our data, expected to decrease for closer stacking since electronic coupling enhances the size homogeneity of the electronic system [13]. But if the layer distance becomes very small, the strain profile in the InAs/GaAs stack can change drastically. Variations in this strain profile between stacks, or even between different dots in one stack, can cause variations in the dot size and confinement energy, and thus also a broadening of the PL line.

The field dependence for a magnetic field parallel to the growth direction (\( B \parallel z \)), probes the in-plane charge confinement. From Fig. 2(a) it is clear that structures A to C behave very much alike for this field direction. In particular the exciton effective mass is constant at 0.13 \( m_0 \) (Table 1; \( m_0 \) is the free electron mass). On the other hand, the structure with the closest stacking, D, shows a larger diamagnetic energy shift due to a smaller exciton effective mass (0.08 \( m_0 \)). Atomic force microscopy on a structure similar to A, but uncapped, reveals an average dot diameter of 15 nm and a height of 1.5 nm [1]. If the dots in our structures have similar heights, then, in structure D with \( d = 3.1 \) nm, then the thickness of the GaAs spacer layer is comparable to the thickness of the dot. Therefore, the capping layer is no longer relaxed. Instead, some strain is introduced, allowing
its partial relaxation in the InAs dot. This reduces the exciton effective mass [6]. The partial strain relaxation inside the InAs dot is confirmed by the Raman shift of the phonon energy, as measured by Ibáñez et al. [14]. They found that the dots in the structure with a capping layer of 5.5 nm are strained as much as those from a single layered structure, while in the structure with a layer spacing of 3.1 nm a decrease in dot strain of 26% was observed.

Variations in the strain profile from stack to stack, or between dots in one stack, can cause a large dot-size inhomogeneity and a broadening of the PL. Also, the large strain in the thin GaAs layer may induce defects in the lattice [15] having the same effect on the line width. Indeed, although the line width already goes up from 35 to 44 meV if \(d\) is reduced from 9.8 to 5.5 nm; the largest increase is observed in coincidence with the strain relaxation: the line width is 68 meV for \(d = 3.1\) nm. This strain relaxation does not have a noticeable influence on the diameter of the dots since the wave function extent does not vary much (Table 1).

When a magnetic field is applied in the plane of the quantum dots (\(B \parallel z\)), the energy shift is influenced by both the height and the diameter of the dots. For structure B, as well as for reference structure A, the diamagnetic shift is small and no linear field dependence is seen in the available field range (Fig. 2). As a result, no absolute values for the exciton radius or effective mass could be obtained. However, the non-linearity in the field range up to 50 T indicates that the exciton radius must be smaller than \(2^{1/2}\times\) times the magnetic length at 50 T, i.e. <5.1 nm. It is therefore clear that for an interlayer distance of 9.8 nm, the charge carriers are not coupled between the different layers in the stack, and that each layer behaves just like a single layer. When the interlayer distance is reduced to 5.5 nm, the diamagnetic shift between 0 and 50 T is doubled from 10.9 to 20.8 meV (Table 1). Since for \(B \parallel z\) no change in the field dependence of the PL is observed, the dramatic increase in shift for \(B \perp z\) must be due to an increase in exciton radius in the growth direction. This is clear evidence that the dots in the stack are electronically coupled. The driving mechanism for hole coupling would be a change in strain in which the dots would relax while the strain in the GaAs spacer layer increases [6]. This would imply a change in mass. Since this is not observed at the onset of coupling (\(d = 5.5\) nm), it must be the electron and not the hole that is

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\begin{array}{|c|c|c|c|c|c|c|c|}
\hline
\text{Sample} & \text{Stack height} & \text{Line width} & \text{Mass} & \text{Radius} & \Delta E & \text{Mass} & \Delta E \\
\text{(nm)} & \text{(meV)} & (B \parallel z) & (B \parallel z) & (B \parallel z) & (B \perp z) & (B \perp z) \text{ (meV)} & \text{(nm)} \text{ (meV)} \text{ (nm)} \text{ (meV)} \\
\hline
\text{A} & \text{(single)} & 1.5 & 35 & 0.13 & 8.9 & 18.9 & <5.1 & 7.3 \\
B & (9.8 \text{ nm}) & 89.7 & 37 & 0.13 & 8.5 & 18.7 & <5.1 & 10.9 \\
C & (5.5 \text{ nm}) & 51.0 & 44 & 0.13 & 7.9 & 18.2 & 0.12 & 9.1 & 20.8 \\
D & (3.1 \text{ nm}) & 29.4 & 68 & 0.08 & 8.8 & 28.8 & 0.08 & 9.9 & 32.9 \\
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\end{array}
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Fig. 2. Diamagnetic energy shift for (a) \(B \parallel z\) and (b) \(B \perp z\). The vertical line in (a) denotes the average cross-over field for \(B \parallel z\) and all structures at \(B = (18 \pm 1)\) T. The arrows in (b) indicate the cross-over field for structures C and D.
coupled over the stack. This is confirmed by recent calculations [5]. For $d = 3.1$ nm it is possible that the strain relaxation in the dot causes also the hole wave function to spread through the stack, but we have no evidence for or against this in our data.

For structure C, the diamagnetic shift for $B \perp z$ is linear from 21 T on, corresponding to an experimental exciton radius of 9.1 nm. This is indeed much larger than the dot height, indicating that the electronic coupling is present. However, this exciton radius is still much smaller than the stack height (51 nm), and is similar to the value obtained for $B \parallel z$ (7.9 nm). In fact, the entire behaviour of this sample is very similar for both field directions, including the exciton effective mass, whereas in general this is not the case [7,8]. Also in structure D, the diamagnetic shifts for both field directions are similar, and again, the stack height (29 nm) is larger than the diameter of the stack. By going from $B \parallel z$ to $B \perp z$, the extent of the confining potential in one of the probed directions increases from 15 nm (dot diameter) to 51 nm for structure C and 29 nm for structure D (stack height), i.e. a tripling and doubling for C and D, respectively. The experimental exciton extent on the other hand increases by only 15% and 13%, and has almost the same value in both structures. This indicates that for $B \perp z$ the diamagnetic shift is determined by a confining dimension that is the same in both structures, i.e. the diameter of the dots, and that it is the strongest confining direction which sets the length scale that determines the diamagnetic shift.

4. Conclusions

We studied ten-fold stacked layers of InAs/GaAs quantum dots by PL in pulsed magnetic fields up to 50 T. The structure with an interlayer distance $d$ of 9.8 nm behaves very much like the single layered reference structure. When $d$ is reduced to 5.5 nm, an increase in the diamagnetic shift for a magnetic field perpendicular to the [0 0 1] direction indicates the onset of electron coupling between the dots in the stack. On reducing $d$ to 3.1 nm a lower exciton effective mass is seen in addition to the coupling. For $d$ this small, the thickness of the GaAs layer in between two successive dots is comparable to the dot height. As a result, the thin GaAs layer is more strained, while the strain in the InAs dot is partially relaxed, reducing the exciton effective mass.

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