Temperature Dependence of the Photoluminescence of Self-Assembled InAs/GaAs Quantum Dots Studied in High Magnetic Fields

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We have investigated the photoluminescence (PL) of self-assembled InAs/GaAs quantum dots (QDs) in high magnetic fields of up to 50 T and as a function of temperature. Our data clearly indicate that two different mechanisms are at work. At low temperatures (T < 80 K), the zero-field PL is increasingly dominated by lower energy dots. High-field measurements however demonstrate that these dots are larger in size only in the growth direction. At temperatures above 100 K, a strong decrease of the PL peak energy shift with field is observed, while the zero-field PL is characterized by a redshift according to the changes in the bandgap. We discuss these contradictory observations in terms of a phenomenon that we call field-assisted enhancement of the QD barrier potential. Since this effect is much stronger for small high-energy QDs, the latter progressively dominate the PL emission when temperature and magnetic field are increased.

Self-assembled quantum dot (QD) semiconductor nanostructures are currently attracting substantial interest both for their fundamental properties and for technological applications.[1-3] For devices like lasers, the occupancy of the discrete atom-like energy levels[4] of the QDs is ideally temperature (T) independent for $T \leq 300$ K, but such ideal behavior is not observed in practice. Hence, a thorough understanding of the effect of temperature is essential for device fabrication. In this work, we have performed nonresonant photoluminescence (PL) studies of InAs/GaAs QDs as a function of temperature in high magnetic fields.[5] Both the zero-field and high field experiments illustrate that two different temperature regimes can clearly be distinguished. At low temperatures ($T < 80$ K), the zero-field PL energy shows a redshift with increasing temperature that is faster than predicted for the InAs band gap by the empirical Varshni law.[6] Correspondingly, high field measurements confirm an enhanced transfer of carriers toward QDs that are larger in size in the growth direction z as the PL peak energy shift (ΔE) with magnetic field applied normal to the growth direction exhibits a clear increase with increasing temperature. However, as there is no such trend for B // z, these low-energy dots are larger only in the growth direction and the exciton extent in the plane of the sample remains unchanged for $T < 80$ K. At higher temperatures ($T > 100$ K) our results show that the zero-field PL energy decreases according to the changes in the InAs band gap, with no sign of change in confinement. The magneto-PL measurements however reveal a dramatic decrease of ΔE that is irrespective of the orientation of the applied magnetic field, implying a shrinkage of the exciton wave-function. We resolve this apparent contradiction by noting that the energy shift ΔE is measured in the presence of a very strong magnetic field, and attribute the increased importance of small high-energy dots for the PL at high fields and temperatures to a field-induced increase of the QD barrier potential.

The sample was prepared using solid-source molecular beam epitaxy. A 0.5 μm thick GaAs-layer was grown on top of a semi-insulating GaAs substrate. Subsequently, 1.8 monolayers of InAs were deposited at a temperature of 480°C. During this phase, QDs were formed following the Stranski-Krastanow growth mode. Finally, the sample was capped with 25.2 nm of GaAs. The sample was excited by the light from a solid-state laser at 532 nm via a 200 μm core optical fiber with a power density of 1 – 10 Wcm^{-2}. The sample itself was located in the bore of a home-made pulsed field coil, placed in a He flow system allowing PL measurements over a broad temperature range (4.2 – 160 K). The coil was connected to a 5 kV 500 kJ capacitor bank, enabling magnetic field pulses of 20 ms and up to 50 T. The PL signal was collected by a single 550-μm core collection fiber and subsequently analyzed by a 30 cm monochromator and a liquid nitrogen cooled InGaAs diode array, using count times up to 4 ms. The entire data set for the complete magnetic field range between 0 and 50 T was then fitted using an excitonic model describing the dependence of the PL energy on magnetic field,[7] whose fitting parameters give access to the exciton effective mass μ and radius $\left\langle \rho^2 \right\rangle^{1/2}$:

$$\begin{aligned} E_{cm} &= E_0 + \frac{e^2 \left\langle \rho^2 \right\rangle}{8\mu} B^2 \qquad \text{for } B < \frac{2\mathrm{h}}{e\left\langle \rho^2 \right\rangle} \\ E_{cm} &= E_0 - \frac{\mathrm{h}^2}{2\mu\left\langle \rho^2 \right\rangle} + \frac{e\mathrm{h}}{2\mu} B \qquad \text{for } B > \frac{2\mathrm{h}}{e\left\langle \rho^2 \right\rangle} \end{aligned} \tag{1}$$

where E_{cm} is the centre of mass PL energy, E_0 is the zero-field PL energy and $\hbar$ is the reduced Planck constant. Through the combination of the parabolic (low fields) and linear (high fields) behaviour of the PL shift into one equation, where the crossover field is given by $B_c = \frac{2\mathrm{h}}{e\left\langle \rho^2 \right\rangle}$, we are able to extract both the effective exciton radius

$\langle \rho^2 \rangle^{1/2}$ and the reduced exciton mass μ, two of the key parameters that govern the opto-electronic properties of semiconductor nanostructures. For the purposes of this investigation, we will concentrate on the shift in energy of the PL peak between 0 and 50 T, ΔE, and the exciton radius, but not the exciton mass.

The inset of Fig. 1 shows the behaviour of the PL peak energy as a function of temperature in the absence of a magnetic field. At low temperatures ($T < 80$ K) the redshift of the PL energy is faster than predicted[6] for the InAs band gap. As discussed previously by a number of groups[8-10], this effect is attributed to thermal excitation of carriers from small high-energy dots to large low-energy dots via the wetting layer. At higher temperatures, the PL energy nicely follows the changes according to the Varshni law for the InAs bandgap. Following Brusaferri et al.,[8] who reported evidence of families of dots with 'remarkably uniform sizes', we interpret this temperature dependence of the PL energy as suggesting the presence of different dot families with distinct sizes in our sample.

In order to gain further insight into the confinement of the carriers inside the dots, we then studied the magneto-PL of the sample at different temperatures and with different orientations of the field with respect to the growth direction. For the low temperature regime, the shift is more or less constant with a magnetic field applied in the growth direction, while experiments with $B \perp z$ reveal an increase of the shift with temperature. When $B \perp z$, we are probing the vertical extent of the exciton, since this is the direction with the strongest confinement with this field orientation. Because the exciton wave-function is so small in this direction, ΔE is dominated by the low field regime where it is proportional to $\langle \rho^2 \rangle / \mu$. An increase in ΔE is thus either because μ is decreasing or $\langle \rho^2 \rangle^{1/2}$ is increasing (or both). However, if it were

only because μ was decreasing this would also increase the confinement energy, and result in an increase of the zero-field PL energy relative to the change expected from the Varshni law, which is the opposite of what we observe. On the other hand, a decrease in the zero-field PL energy is entirely consistent with an enhanced transfer of carriers towards larger QDs leading to a decrease in confinement. Hence, the increase in ΔE with the magnetic field perpendicular to the growth direction indicates an expansion of the exciton wave-function in the growth direction reflecting the increased contribution of larger QDs to the PL emission. In contrast, the energy shift with magnetic field parallel to the growth direction remains unaltered at temperatures below 80 K. Thus, the explanation of the data in the inset of Fig. 1 that there is thermal excitation from smaller to bigger dots is indeed supported by our magneto-PL measurements, but the magneto-PL data crucially reveal that *these dots are larger in size only in the growth direction*: there is no indication of a change in the average lateral size of the ensemble of dots in this temperature range.

We now turn to the high temperature regime. Here, the field-induced PL shift is characterized by a massive decrease as a function of temperature irrespective of the B orientation (Fig 2.). Applying an analysis to the data following the magnetic field dependence of the peak energy as described in Eq. (1) would lead us to interpret this as a shrinkage of the exciton radius (or, equivalently, an increase in confinement) as a result of increasing temperature (inset of Fig. 2). However, if the QDs active at higher temperatures were, on average, smaller than the ones responsible for the PL at low temperatures, their confinement energy should be higher, as would be the corresponding transition energies within the QD. In this case, the zero-field PL energy would not follow the empirical Varshni law for the InAs bandgap, but show a weaker temperature dependence or even an increase as a function of temperature, i.e. the

exact opposite of what we observe at low *T*. However, this is clearly not the case, and we therefore conclude that the decrease of the field-induced PL shift with temperature is not due to a decrease of the (zero-field) exciton radius as a function of temperature, but that it has some other cause.

The contradiction is resolved when noting that the energy shift is determined in the presence of a very strong magnetic field, which, we propose, affects the confinement potentials. In general, applying a magnetic field causes the energy levels of bulk GaAs (the barrier material) to be lifted by $\frac{1}{2}\hbar\omega_c$, or about 0.9 meVT^{-1}. In contrast, the PL energy shift between 0 and 50 T of the InAs QDs with *B // z* for the considered temperature range, varies between 10 and 20 meV (see Fig. 2), resulting in an energy shift of about 0.2 to 0.4 meVT^{-1}. Fig. 3 shows a schematic representation of the electron energy levels of an InAs QD in the GaAs matrix, and illustrates the fact that the application of a strong magnetic field increases the energy gap between the states in the QDs and the states of the surrounding bulk GaAs material. When applying a magnetic field, the energy levels of the GaAs barrier material are substantially raised, while the increase of the QD levels is rather moderate. Consequently, a strong magnetic field enhances the temperature stability of the system by counteracting the escape of carriers from the dots into the surrounding barrier material. However, the influence of a magnetic field on the energy levels of the QDs is strongly dependent on the degree of confinement present in the dots. Smaller dots have a stronger confinement potential, resulting in a weaker dependence on magnetic field. Therefore, the energy levels of the excitons with *strong lateral confinement*, i.e. those with a weak energy shift in magnetic field (~ 0.2 meVT^{-1}), are *lifted much less* than the ones with a weak lateral confinement, as the latter have a larger PL shift in field (~ 0.4 meVT^{-1}). In both cases, however, the barrier energy is

raised by the same larger amount. This results in a field-induced enhancement of 0.7 to 0.5 meVT^{-1} of the QD barrier potential. Correspondingly, the extra energy difference between the QD levels and the GaAs barrier material at 50 T is 35 meV (400 K) for the high energy dots (i.e. the ones with strong lateral confinement), while it is only 25 meV (290 K) for the low energy dots (i.e. with weaker lateral confinement). In other words, the effect of the magnetic field is to deepen the confinement potentials of the QDs, and counter-intuitively, the mechanism is much stronger for smaller, high energy dots, as the energy levels of these dots are the least affected by a magnetic field. The result is that at high temperatures and high magnetic fields the contribution of the high energy dots to the PL relative to the low energy dots is increased, which in turn results in a smaller PL shift in magnetic field.

In conclusion, we have studied exciton confinement in self-assembled InAs/GaAs QDs. At low temperatures (T < 80 K), we find that there is thermal excitation via the wetting layer to dots which are larger *only* in the growth direction. At higher temperatures (T > 100 K) we observe a strong decrease of the PL shift in field, which we attribute to field enhancement of the QD barrier potential. As the influence of a magnetic field on the energy levels of the QDs depends inversely on the confinement energy, this mechanism is particularly important for smaller dots.

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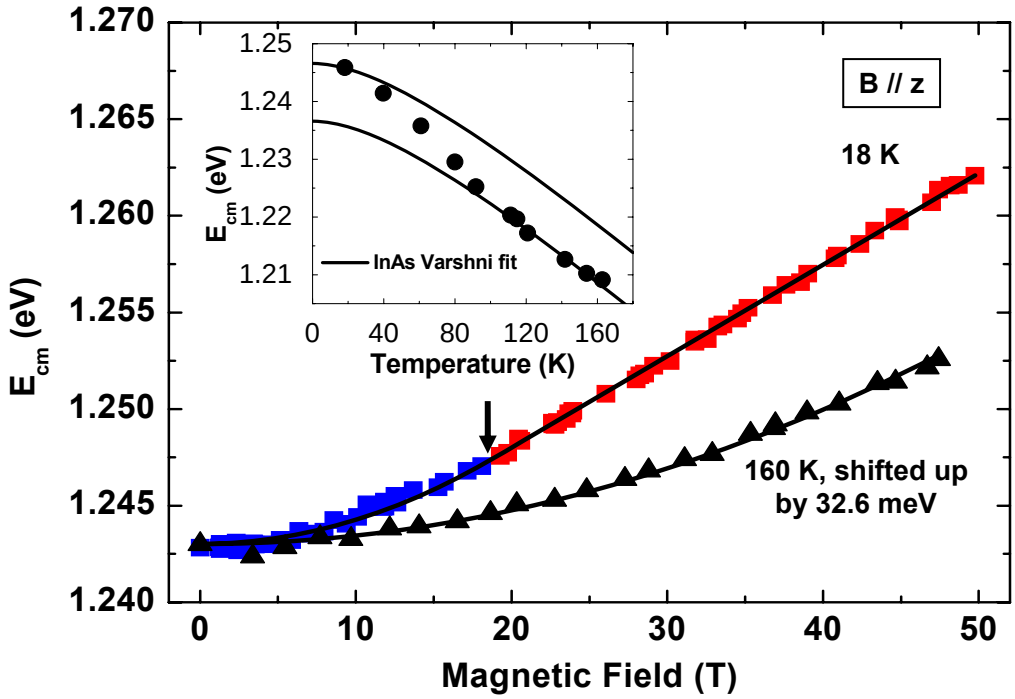

Figure 1 (color online)**:** The PL energy, E_{cm}, versus magnetic field. The data for 160 K has been shifted for clarity. The lines are fits to the data using Eq. (1). At lower temperatures, a crossover from parabolic to linear behavior occurs at 19 T (indicated by the arrow), whereas the 160 K data is parabolic up to 50 T. Note that the energy shift is much larger for low temperatures. The inset shows the zero-field PL energy as a function of temperature. For low temperatures, the decrease of the PL energy with temperature is faster than predicted by the Varshni law[6] for the InAs bandgap (full lines).

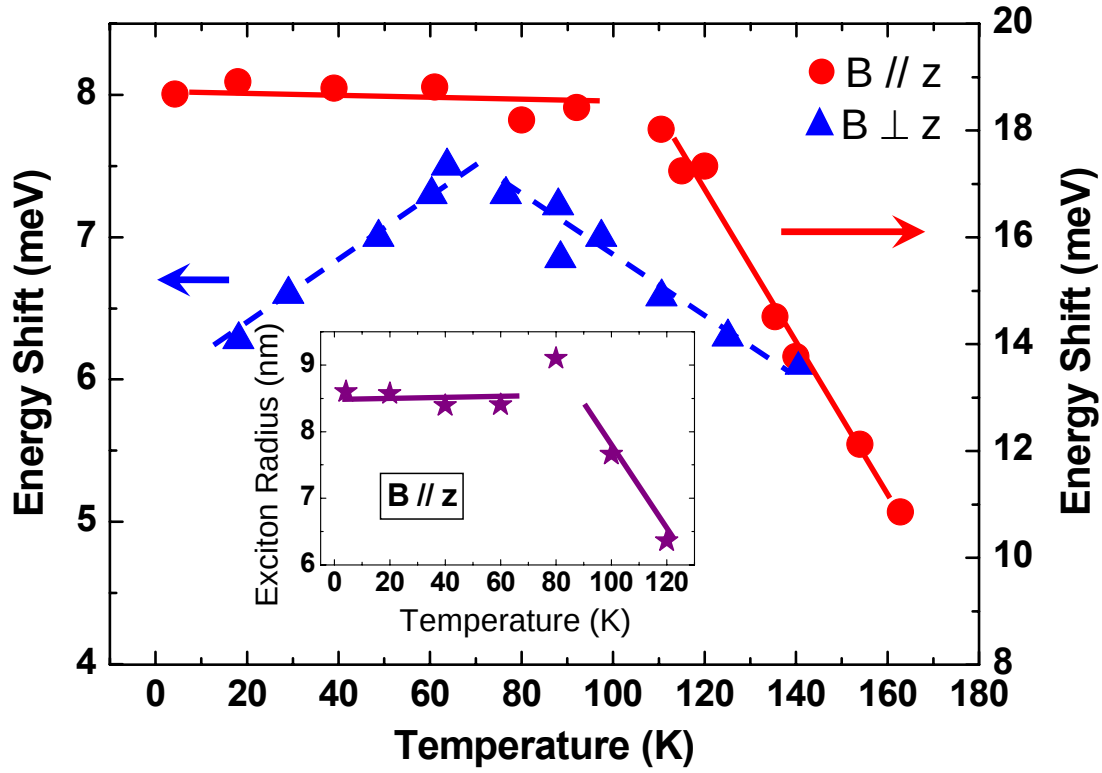

Figure 2 (color online)**:** Energy shift between 0 and 50 T versus temperature for a magnetic field applied parallel (circles) and perpendicular to the growth direction (triangles). A strong decrease of the PL shift characterizes the high temperature ($T > 100$ K) regime. The inset shows the exciton wave-function extent as a function of temperature following Eq. (1) for $B // z$, and reveals an apparent wave-function shrinkage at high temperatures. For $T > 120$ K the lateral exciton radius becomes so small that the cross-over to the high-field regime is at too high a field to be determined reliably. The lines are guides to the eye.

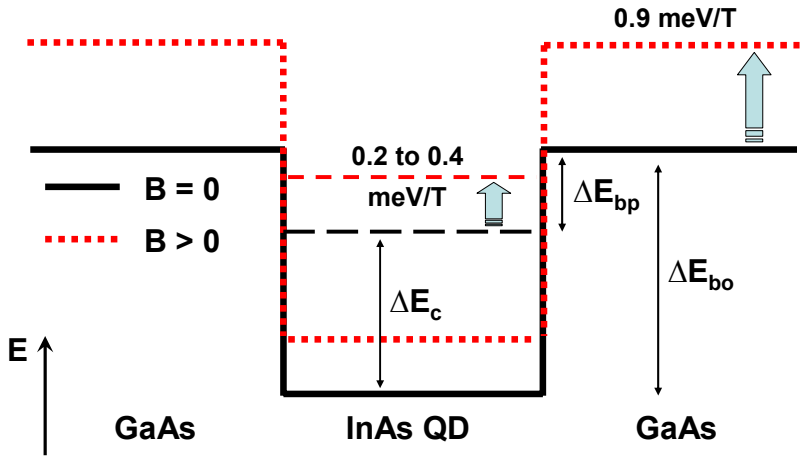

Figure 3 (color online)**:** Schematic representation of the electron energy levels of the InAs QDs and GaAs matrix material as they are lifted by the magnetic field. The differential rise in energy levels as a function of B increases the effective barrier height by 0.5 to 0.7 meVT^{-1}. The confinement energy ΔE_c, the barrier potential ΔE_{bp} and the band offset ΔE_{bo} are indicated.