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Exciton fine structure splitting in InP quantum dots in GaInP

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Abstract

We have investigated the electronic structure of excitons in InP quantum dots in GaInP. The exciton is theoretically expected to have four states. Two of the states are allowed to optically decay to the ground (vacuum) state in the dipole approximation. We see these two lines in photoluminescence (PL) experiments and find that the splitting between the lines (the fine structure splitting) is 150($\pm$30) μeV. The lines were perpendicularly polarized. We verified that the lines arise from neutral excitons by using correlation spectroscopy. The theoretical calculations show that the polarization of the emission lines are along and perpendicular to the major axis of elongated dots. The fine structure splitting depends on the degree of elongation of the dots and is close to zero for dots of cylindrical symmetry, despite the influence of the piezoelectric polarization, which is included in the calculation.

1. Introduction

Most previous studies on single dots have been performed on InGaAs quantum dots in GaAs. We have here investigated InP quantum dots in GaInP in order to check whether the results on InGaAs quantum dots can be generalized to this system. Single-dot polarized photoluminescence spectroscopy, correlation spectroscopy and theoretical calculations have been performed. We find good agreement between experiment and theory and we can conclude that the InP/GaInP system behaves in a similar way to the InGaAs/GaAs system.

Excitons in quantum dots based on direct gap III–V semiconductors have four energy levels as shown theoretically and experimentally [1, 2]. These levels appear in two doublets which are split by the exchange interaction, which is shown in figure 1. This splitting due to the exchange interaction is debated in the literature and values ranging from 0.1 meV [3] to 5 meV [1] have been reported and seems to depend on the materials system investigated. We will use the value of 5 meV in this paper although the exact value is of minor importance in this investigation. Two of the levels are forbidden to decay optically and two are allowed. The two allowed lines

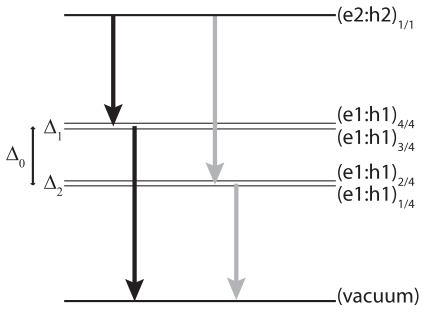

Figure 1. The four exciton states and the biexciton state in a quantum dot. The two lower exciton states are dark, as indicated by the grey arrows. The higher pair is bright and the transitions are indicated by black arrows. Δ_0 is the splitting between the dark and the bright states. Δ_1 is the splitting between the two bright exciton states, the fine structure splitting.

are split by a small amount, typically 100 μeV—this is referred to as the fine structure splitting and is denoted Δ_1 in figure 1. It is the higher energy doublet which is optically allowed, and is called the bright state, while the lower energy doublet is forbidden and is called the dark state. The dark state is forbidden since a spin flip is required when a transition to the ground, i.e. the vacuum state, occurs. Earlier studies, most of all of InAs quantum dots (QDs) embedded in GaAs, have demonstrated that it is important to have a very small fine structure splitting if QDs are used to produce entangled photon pairs [4, 5]. These experiments have also shown that the fine structure splitting can be greatly reduced, even approaching zero, by manipulating the geometry of the dot [6]. It is often argued that the potential of a quantum dot can have cylindrical symmetry by choosing the right growth conditions and the amount of annealing [7], but this can only be true if the piezoelectric polarization is not taken into account. Consider that a strained QD in III–V materials experiences a piezoelectric polarization which is strongest at the interfaces and that the sign of the piezoelectric polarization is different for, say, the 111A and the 111B interfaces. It is then clear that complete cylindrical symmetry of the potential cannot be obtained even in a dot where the geometry is of cylindrical symmetry.

We use a notation for the energy levels that does not make any assumptions about the symmetry of the QD. This notation is thoroughly described in Landin *et al* [1]. The notation is based on energy considerations and avoids any references to atomic physics notation which, due to historical reasons, is very cumbersome. In this notation, the biexciton is labelled $(e2{:}h2)_{1/1}$, the two 'dark' excitons are labelled $(e1{:}h1)_{1/4}$, and $(e1{:}h1)_{2/4}$ and the two 'bright' excitons $(e1{:}h1)_{3/4}$. and $(e1{:}h1)_{4/4}$. The state without excitons is labelled (e0:h0). The energy level diagram of an exciton is shown in figure 1.

2. Experiment

The InP QDs were grown by metal–organic vapour phase epitaxy (MOVPE) in the Stranski–Krastanov mode on GaAs substrates. After a GaAs buffer layer was grown, 300 nm $Ga_{0.52}In_{0.48}P$ was deposited, which is lattice matched to GaAs. A few monolayers of GaP were grown before two monolayers of InP was deposited. The sample was then annealed for 12 s, when the dots were formed, and subsequently overgrown by 200 nm of $Ga_{0.52}In_{0.48}P$. Our dots are situated on a wetting layer of about one monolayer of InP. The growth steps are the same as in [8]. The barrier material is weakly n-type.

The dots are either fully grown [9] or much smaller. That is, we have a bimodal growth. The fully grown dots are 40 nm wide and 15 nm high and emit at roughly 1.65 eV. They are

shaped like truncated pyramids with a hexagonal base. The smaller dots are about 20 nm wide and 2–3 nm high [8] with a quite large variation in the lateral size. The small dots emit from 1.8 eV up to the energy of the wetting layer and the $Ga_{0.52}In_{0.48}P$ at 1.9 eV. The ground state in the fully grown QDs is below the Fermi level, making the dot negatively charged. Smaller dots, such as those investigated here, are neutral however, since the confinement pushes the ground state energy above the Fermi level. We have measured photoluminescence spectra from tens of QDs of the smaller kind on this sample.

The sample was mounted on a cold finger in a cryostat and the photoluminescence was measured at 10 K in the backscattering mode. For excitation we used a frequency-doubled YAG laser emitting at 532 nm. The laser beam made an angle of about 45% with respect to the normal of the sample. The laser spot was typically 0.2 mm in diameter in order to find point-like scatterers for position tracking. The excitation power was varied from 2 mW cm^{-2} to 2 W cm^{-2} for the nonpolarized spectra, fixed at 0.7 W cm^{-2} for the correlation measurements and at 0.2 W cm^{-2} for the polarized spectra. We used an external 20× objective with a numerical aperture of 0.4 to collect the emitted light from the sample. The emitted light was dispersed by a monochromator and detected by a CCD camera for the photoluminescence measurements.

The correlation measurements are time-consuming and it is necessary to stabilize the sample position. We used a positioning system in a closed loop to achieve micrometre stability. The position on the sample was tracked by scattered laser light seen through the back of a dielectric mirror. We could then easily integrate for hours. The emission from one QD was focused on a 20 μm pinhole. The light was then split by a nonpolarizing beam splitter and in each arm refracted by holographic gratings working in transmission. The 50 μm exit slits for the two monochromators were mounted directly on the apertures of Si avalanche photo diodes (APDs) which are single-photon detectors. The spectral resolution of the system was 0.6 nm. The signals from the APDs were correlated in time by a correlation card. This means that one APD acts as a trigger and starts a clock when detecting a photon. The second APD stops the clock when detecting a photon. It is thus possible to measure the statistics of the time delay of the emitted photons in the two arms of the interferometer. Due to the gratings we can freely choose the energy of the photons as well and perform cross-correlation measurements. The time resolution of the whole system was measured to be 0.8 ns. A more detailed description of such a Hanbury-Brown and Twiss interferometer can be found in for example [10].

In figure 2 we show the excitation power dependence of the PL emission from one quantum dot. There are two main lines denoted X and X_2, corresponding to the exciton and the biexciton, respectively. The X line can be seen to be a doublet. With increasing excitation power, the PL intensity of the exciton line should increase linearly, while the intensity of the biexciton line should increase quadratically before saturation, in a first approximation. This was also observed as seen in figure 2. This simple prediction can be made much more refined by solving the rate equations [11].

It is well known that QDs emit photons in cascades [12, 13]. For example, one transition cascade is $(e2{:}h2)_{1/1} \rightarrow (e1{:}h1)_{3/4} \rightarrow (e0{:}h0)$. This means in words, that a biexciton recombines and forms an exciton, which subsequently recombines to the vacuum state (a dot without excitons). On a timescale determined by the exciton recombination time, we expect an increased probability (bunching) of measuring a photon from the biexciton before registering a photon from the exciton and a decreased probability (anti-bunching) of observing the opposite case.

Thus, we can distinguish the exciton and biexciton lines by cross-correlation measurements [12]. This is important since intensity dependent PL is not enough to give a definite assignment of the observed emission lines. In figure 3, we show the results of the correlation measurement. We have set a nominal time, $t = 0$ to the crossover time from

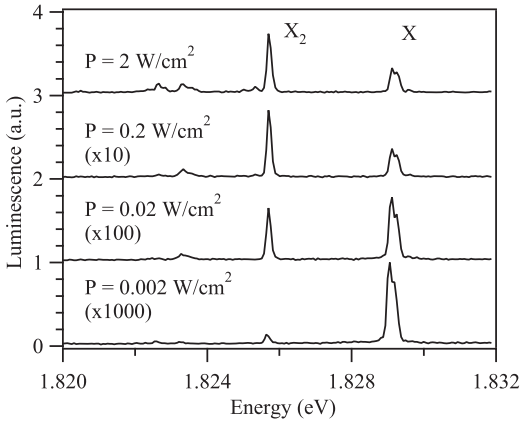

Figure 2. Photoluminescence spectra of the quantum dot at four different excitation intensities. The double peak to the right is the exciton (X) doublet, while the single peak at about 1.826 eV is the biexciton (X_2) line.

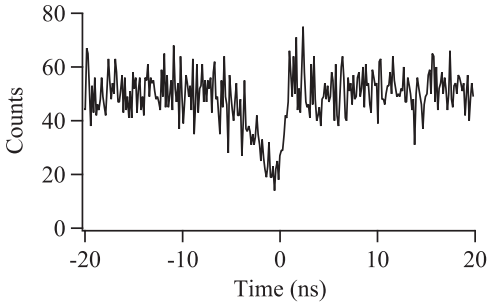

Figure 3. Cross-correlation of photons from excitons and biexcitons. Time is the delay between a start event from a photon originating from the biexciton and a stop event from a photon originating from an exciton in the quantum dot. At positive times we see a peak and at negative times a dip, meaning that the biexciton emission precedes the exciton emission.

bunching to anti-bunching. For all negative times, a photon originating from an exciton is registered before a photon from a biexciton. For positive times the reverse holds. As expected, there is a clear asymmetry. We observe anti-bunching for small negative times and some bunching for small positive times. This indicates that one peak is due to a biexciton and one to an exciton. Note in particular that one of the lines cannot be a charged exciton (biexciton) and the second a neutral biexciton (exciton). Neither can one peak be a charged exciton and the second a neutral exciton. There should in these cases be no correlation between the peaks. It is still possible that the peaks are due to a charged exciton and a charged biexciton [14]. This possibility can be excluded by the following argument. A charged exciton as well as a charged biexciton has only one emission line, and as seen in figure 2 we observe two lines from the exciton. We thus confirm our identification of the emission lines shown in figure 2 by the correlation experiment. This observation is crucial because it is known that charged and neutral excitons can simultaneously be seen in the emission of single quantum dots.

The polarization resolved PL measurements were made with a polarizing prism and a $\lambda/2$-plate inserted between the microscope and the spectrometer. The polarizer was rotated to maximize the grating efficiency of the spectrometer. The wave plate was then rotated in steps

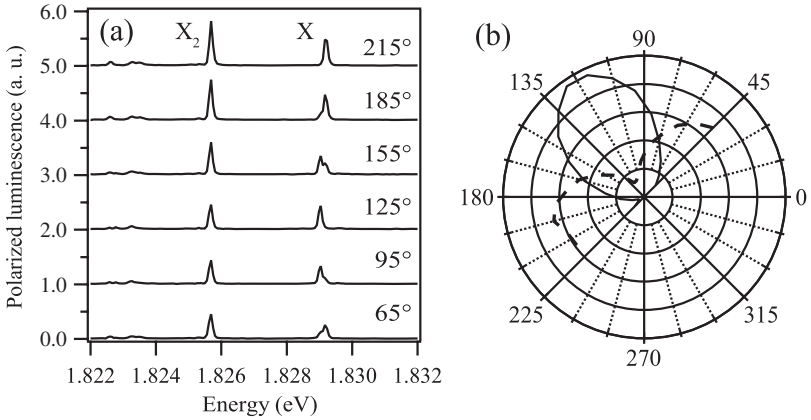

Figure 4. (a) Polarized photo luminescence spectra for six different linear polarization directions. The angles are relative to the [110]-axis of the sample. (b) Relative intensity of the two bright exciton lines, measured at 19 different polarization angles. The solid line corresponds to the exciton state with higher energy.

of 5°. We found that the lower energy peak, $(e1{:}h1)_{3/4} \rightarrow (e0{:}h0)$, has its maximum at about 120° from the [110]-axis and the higher energy peak, $(e1{:}h1)_{4/4} \rightarrow (e0{:}h0)$, is polarized 30° from the [110]-axis, as shown in figure 4(b). The two peaks are thus polarized in orthogonal directions.

The energy split between $(e1{:}h1)_{3/4}$ and $(e1{:}h1)_{4/4}$ is 150($\pm$30) μeV, as seen in figure 4(a). The splitting of the biexciton line is much smaller, indeed smaller than 100 μeV, which is the resolution of our spectrometer. From figure 1 it can be seen that the emission from the exciton and the biexciton should be mirror images of each other since the splitting occurs in the exciton state, with both the vacuum and the biexciton consisting of a single state. It is thus very puzzling that we observe the fine structure only in the exciton line and not in the biexciton line. Note that the assignment of the lines is very certain from the correlation experiment and the discussion above and the natural explanation that the putative biexciton is in fact a charged exciton does not hold. Our tentative explanation is that the transition probabilities for the transition between the biexciton and the exciton strongly favours only one of the final states in the exciton. This observation is not born out by theory and neither is it expected from simpler arguments. It is a thus an unsolved problem. The other QDs in this sample have shown significantly smaller fine structure splitting.

3. Theory

We have calculated the energy structure of the exciton in three steps. First we calculated the strain field using a three-dimensional continuum model using linear elasticity theory [15]. It has been shown that this model is very accurate and is also in agreement with the valence force model which is more atomistic. The strain was minimized using a conjugate gradient algorithm on a cubic grid of 180·180·90 sites. Since the strain field decays as a power law it was necessary to use a large grid. Using this strain field we calculated the single-particle states using an eight-band $\mathbf{k}\cdot\mathbf{p}$ model. The Hamiltonian was discretized on the same cubic grid as used for the strain. The grid in this case was truncated about a factor of eight in volume, compared with the initial volume, since the electronic states decay exponentially in the barrier material. The resulting matrix is very sparse and was diagonalized using the Lanczos algorithm, which is efficient

for sparse systems. For these two stages we used a computing time of about 2 h on a personal computer. The single-particle states were then used to find the exciton levels where we included the direct and exchange interaction between the electron and hole [16]. The parameters for the strain calculations as well as for the electronic structure calculations were taken from a review by Vurgaftman *et al* [17]. The exchange interaction parameter is not known, however, and we used a value which has been deduced from experiments on InAs quantum dots in GaAs [1] although the exact value is not important here since we study the bright state.

We found a splitting between the doublets of about 4.5 meV and a splitting between $(e1{:}h1)_{3/4}$ and $(e1{:}h1)_{4/4}$ of about 20 μeV. When we calculated the polarization dependent transition matrix elements we found that the two levels are polarized in orthogonal directions, along [110] and along $[1\bar{1}0]$. We thus confirm the main features of our experiment, the energy split between $(e1{:}h1)_{3/4}$ and $(e1{:}h1)_{4/4}$ and the observation that they are polarized in orthogonal direction. However the direction of polarization is slightly different. We attribute this to the unknown details of the shape of our dot. The calculations use a dot that is elongated along [110] which may not correspond to our measured dot. In order to check this, we calculated the polarization dependent transition matrix elements for a dot which was elongated along [100] where we find that the two levels are polarized along [100] and [010]. We thus find that the polarization axes of the emission lines are parallel and perpendicular to the main axes of an elongated dot. Concerning the fine structure splitting we also calculated the electronic structure of a dot with cylindrical symmetry, modelled as half sphere. The potential does not have cylindrical symmetry since we include the piezoelectric polarization in our calculations. However, we found that the splitting of the optically allowed states, i.e. Δ_1, is less than 10 μeV. For a dot having C_{4v}-symmetry we find that $\Delta_1 = 60$ μeV. This dot was modelled as a pyramid bounded by {111} planes. It is thus possible to use the fine structure splitting and the polarization data to obtain information about the shape of the dot.

In summary, we have investigated the fine structure splitting of InP quantum dots in GaInP and found good agreement with eight-band $\mathbf{k} \cdot \mathbf{p}$ theory. We find that polarization dependent measurements along with measurement of the energy split of the allowed states allow some information about the shape of the dot to be derived.

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