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Investigation of interband optical transitions by near-resonant magneto-photoluminescence in InAs/GaAs quantum dots

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Abstract. Resonant photoluminescence experiments performed on self-assembled InAs/GaAs quantum dots under strong magnetic field up to 28 T give rise to an accurate determination of the interband magneto-optical transitions. As this technique minimizes the effect of the homogeneous broadening of the transitions due to the size and composition fluctuations of the dots, the experimental spectra display well-defined peaks. A good agreement is found between the experimental data and calculations using an effective mass model including the coupling between the mixed exciton-LO phonon states. Transitions involving excitonic polarons are clearly identified. Moreover, a light-hole to conduction transition is also evidenced in agreement with previous theoretical predictions.

PACS. 71.38.-k Polarons and electron-phonon interactions – 73.21.La Quantum dots – 78.20.Ls Magnetooptical effects

1 Introduction

The interband optical transitions in semiconductor quantum dots (QDs) have been extensively studied on dot ensembles [1–3] and on single dots [4–6]. The excitonic optical absorption spectra provide useful information on the nature of the confined states, which is crucial both for fundamental physics and for applications, for instance in the field of quantum computing [7]. Moreover, magneto-optical experiments allow a precise assignment of the various observed transitions, as the effect of an applied magnetic field is different for s, p, and d states as well as for conduction and valence band states. Nevertheless, the analysis of excitonic optical spectra is usually intricate. Firstly, excitons in QDs strongly couple with LO-phonons and form the so-called excitonic polarons which lead to significant modifications of the confined transition energies [2,8–10]. Recent experiments using photoluminescence excitation spectroscopy under strong magnetic field have evidenced the existence of the excitonic polarons in self-assembled InAs/GaAs QDs [2]. Secondly, recent calculations of the excitonic optical spectra using an atomistic pseudo-potential approach [11] have suggested that, in addition to a heavy-hole to conduction band transitions, light-hole to conduction band transitions could also be observed as the ground light-hole state is calculated to be weakly confined in InAs/GaAs QDs. Moreover, it is well-known that interband transitions in a QD ensemble are inhomogeneously broadened because of fluctuations of the confining potential which arise from size and composition dispersion. A precise analysis of the interband transitions thus requires overcoming this inhomogeneous broadening by using either photoluminescence excitation (PLE) or resonant photoluminescence (RPL) techniques [12]. The RPL technique is a good choice, in particular, because its spectra does not include the large peak associated with the wetting layer that is found in PLE spectra [2]. Indeed, PLE measurements are affected by the WL states even if the excitation energy is below the ground WL transition, because of the so-called "crossed transitions" [13]. These transitions introduce an absorption tail (and thus an important additional contribution to the PLE signal) for energies well below the WL ground state transition. The main difference between RPL and PLE is that in the latter, the number of crossed pairs changes when changing the laser energy while in the former, it is kept constant. The peaks in the RPL measurement are therefore well defined and more easily identifiable than those of the PLE measurement. In the present work, we have studied at $T = 4$ K the RPL spectra under strong magnetic field up to 28 T of several ensembles of self-assembled InAs/GaAs

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QDs. In a RPL experiment, the excitation energy is less than the fundamental excitation (bandedge) of the wetting layer, so that, for a given energy, only subensembles of similar QDs are resonantly excited and contribute to the PL signal. The subensembles of excited QDs are defined by the various allowed transitions. For each magnetic field, the RPL spectra display well-defined peaks, making low temperature RPL spectroscopy an appropriate probe of the QD confined states. The magnetic field dependence of the peak energies allows for an unambiguous assignment of the interband transitions. The energies and the oscillator strengths of the transitions are calculated using an effective mass model including the coupling between the mixed exciton-LO phonon states and the effects of an applied magnetic field, as described in reference [2]. A good agreement is found between these calculations and the experimental data, evidencing that transitions involving excitonic polarons are observed when two exciton-phonon states have close enough energies with phonon occupations differing by one. Moreover, an additional peak at high energy which could be assigned to a light-hole to conduction transition [11] is observed, showing that the ground light-hole state may be confined in our QDs.

2 Experimental details

The InAs/GaAs self-assembled QDs investigated here were grown on (001) GaAs substrates by molecular beam epitaxy using the Stranski-Krastanov growth mode [14]. Each sample contains a multistack of 20 layers of InAs QDs separated by 50 nm barriers, each layer having a QD density of $\sim 4 \times 10^{10}\,\mathrm{cm}^{-2}$. The InAs QDs were formed by depositing 2.2 monolayers (ML) at 520 °C with a growth rate of 0.12 ML/s. There were no growth interruption before GaAs overgrowth. The shape of the InAs islands, estimated from cross-sectional transmission electron spectroscopy in samples grown in similar conditions, is lenslike with a height of ∼2 nm and a lateral diameter of ∼20 nm. In the following, we discuss in detail results obtained from a sample that contains a Be δ-doping of the GaAs barriers at 2 nm under the dot layers. Similar results found for undoped and n-doped samples will also be presented. The samples were characterized by photoluminescence (PL) and by FIR magneto-spectroscopy at 2 K. The PL peak, in all samples, was centered at 1.2 eV with a *FWHM* of ∼50 meV. An energy separation of ∼47 meV (∼22 meV) between the ground and the first excited conduction (valence) states was measured by FIR magneto-spectroscopy in similar samples. We have performed low temperature ($T = 4$ K) interband measurements on these QD ensembles using the RPL technique under strong magnetic field applied along the sample growth axis. Measurements up to 28 T have been done at the High Magnetic Field Laboratory in Grenoble. The samples were mounted in a resistive magnet and immersed in liquid helium. A Ti:sapphire laser was used. All experiments were conducted at a constant weak excitation power in order to remain in the linear regime. A system of optical fibers were used for the sample excitation and the collection of the PL signal. The emitted light from the sample is dispersed through a Jobin Yvon spectrometer and detected by a photomultiplier.

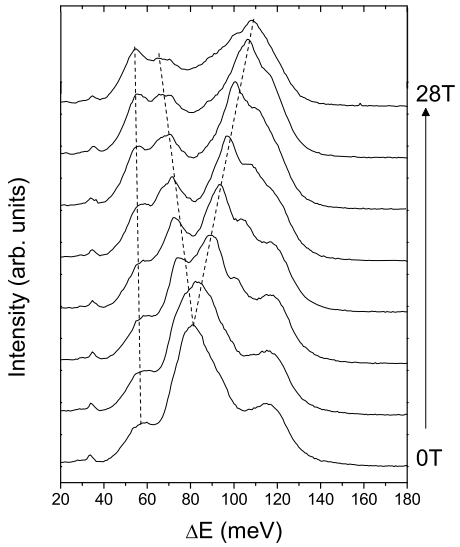

Fig. 1. Magneto-RPL spectra recorded on a p-doped sample at 4 K from $B = 0$ to $B = 28$ T every 4 T and for a $E_{exc} =$ 1293 meV. Traces have been vertically offset for clarity. The dashed lines are guides for the eyes.

3 Results

Interband transitions of a QD ensemble are inhomogeneously broadened because of fluctuations of the confining potential which arise from size and composition dispersion. The broad bell-like form of a PL peak is more or less a measure of size inhomogeneity. In RPL spectroscopy, because the fixed excitation energy is less than the GaAs gap and the InAs wetting layer, only subensembles of similar sized QDs are resonantly excited and contribute to the PL signal. Dots that possess an excited state in resonance with the fixed excitation energy, E_{exc}, are excited. The system relaxes to its ground state and gives off a photon of energy equal to its ground state energy. The detection energy, E_{det}, is varied such that several resonances are observed, each one corresponding to the signal of a subensemble of similar dots. Figure 1 depicts the RPL spectra for an excitation energy $E_{exc} = 1293$ meV recorded at 4 K on a p-doped sample from $B = 0$ to 28 T every 4 T. The x-axis shows the excess excitation energy $\Delta E = E_{exc} - E_{det}$. At 0 T, three distinct features are observed: a low energy peak at ∼59 meV, a high energy peak at ∼116 meV and a more pronounced peak at ∼80 meV. The low energy peak decreases slightly in energy and increases in intensity as the magnetic field is increased. The peak initially at 80 meV splits into two separate peaks: one peak that increases in energy with the magnetic field and one that decreases. In addition, we observe an exchange in intensity between this descending peak and the low energy one. The high energy peak is observed to stay nearly constant in energy with the magnetic field, until it disappears for magnetic fields greater than 16 T. Finally, we note the appearance of a small feature at 8 T

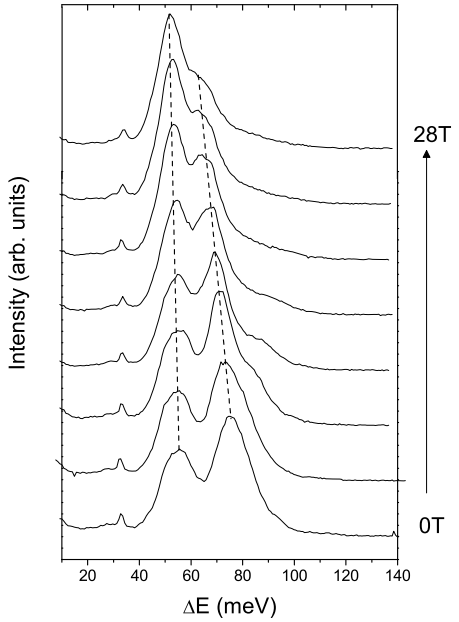

Fig. 2. Magneto-RPL spectra recorded on a p-doped sample at 4 K from $B = 0$ to $B = 28$ T every 4 T and for a $E_{exc} = 1260$ meV. Traces have been vertically offset for clarity. The dashed lines are guides for the eyes.

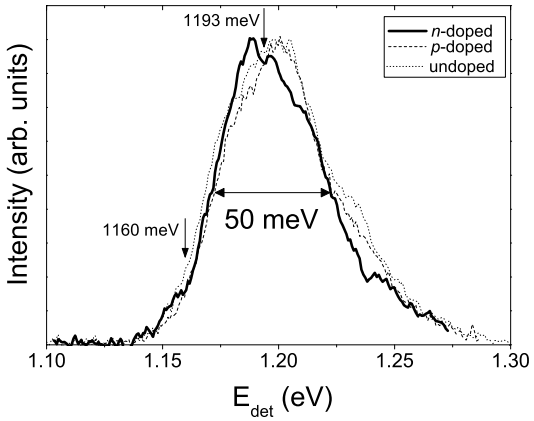

Fig. 3. The normalized PL peaks for three differently doped samples excited by an Ar+ laser and recorded at 4 K.

between the two high energy peaks at ∼101 meV. This peak increases in energy until a magnetic field of 24 T, after which it disappears. We compare this behavior with the same experiment conducted for an excitation at lower energy, $E_{exc} = 1260$ meV as seen in Figure 2. At this excitation energy, only two features are observed at 0 T: a peak at ∼55 meV and a peak of higher energy at ∼75 meV. The two peaks undergo an obvious exchange in intensity as the magnetic field is increased. No features above an energy of 100 meV are observed.

The origin of the differences in the spectra obtained for $E_{exc} = 1293$ meV as compared to $E_{exc} = 1260$ meV is the existence of a distribution of different sized dots in our samples. As noted earlier, the broad bell-like peak of a PL spectrum is a good indication of the size inhomogeneity among the dots in a single sample. Figure 3 shows the PL spectra for three differently doped samples. We observe, for all three samples, a broad peak centered at 1.2 eV with a *FWHM* of ∼50 meV. We can therefore conclude that our samples contain a large population of dots with interband ground state energy equal to 1200 meV. Progressively less dots are found the further the distance from 1200 meV. Finally no dots exist for an interband ground state energy greater than (less than) ∼1280 meV (∼1140 meV). For an excitation energy of 1293 meV the peak at $\Delta E = 100$ meV corresponds to a detection energy, $E_{det} = 1193$ meV. As shown by the arrow in Figure 3, a large population of dots exists at this detection energy and therefore peaks at 100 meV and above are observed in the RPL spectra for $E_{exc} = 1293$ meV. This is not the case for $E_{exc} = 1260$ meV. At this excitation energy, QD states with $\Delta E \geq 100$ meV will have detection energies $E_{det} \leq 1160$ meV. These energies are found in the tail of the PL peak as seen in Figure 3. It follows that the population of QDs possessing such detection energies is very small or non-existent. Therefore peaks above 100 meV, that are found in the RPL spectra for $E_{exc} = 1293$ meV, are not observed for an $E_{exc} = 1260$ meV.

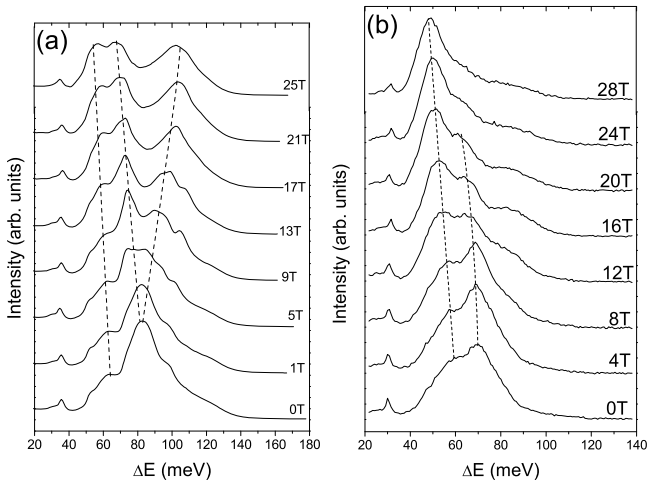

Fig. 4. Magneto-RPL spectra recorded on an undoped sample at 4 K from $B = 0$ to 25 T for $E_{exc} = 1281$ meV (a). Magneto-RPL spectra recorded on a n-doped sample at 4 K from $B = 0$ to 28 T for $E_{exc} = 1259$ meV (b). Traces have been vertically offset for clarity. The dashed lines are guides for the eyes.

Finally, in Figures 4a and 4b we display several selected magneto-RPL spectra for our undoped sample and n-doped sample respectively. The spectra of the undoped sample were taken at the high excitation energy, $E_{exc} = 1281$ meV (Fig. 4a). The results are therefore similar to those obtained in Figure 1. We are able to observed the low energy peak and the peak that splits into two separate peaks with the applied magnetic field. The high energy peak is not detected at this excitation energy for this sample.

The selected RPL spectra of the n-doped sample was taken at a lower excitation energy, $E_{exc} = 1259$ meV (Fig. 4b). These results are therefore comparable to those

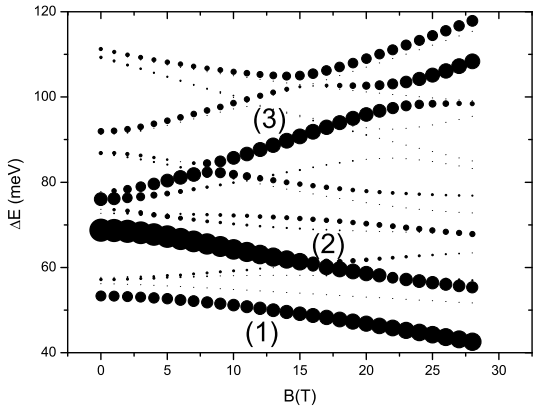

Fig. 5. Calculated excitonic polaron energies and intensities as a function of the magnetic field from reference [2]. The area of the circles are proportional to the oscillator strengths.

of Figure 2 where only two peaks are observed: the low energy peak and the lower branch of the peak that splits into two with the magnetic field. We observe the same excitation energy dependent behavior in the spectra of all of our differently doped samples. In addition, a strong anticrossing between the two low energy peaks is observed in all of our samples. Such a behavior cannot be explained using a purely electronic model. It is necessary to use a model that takes into account the coupling between the optical phonons and the photo-created electron-hole pair in the QD. Such a model is presented in the next section.

4 Analysis and discussion

In spite of their electrical neutrality, excitons have been shown to be in a strong coupling regime with LO-phonons [8]. We have recently reported on the calculation of excitonic polarons under strong magetic field in reference [2]. The calculated polaron energy levels as a function of magnetic field from this reference are presented in Figure 5. The area of the circles in the figure are proportional to the oscillator strengths of the polarons. We have labeled the three polaron states with the largest oscillator strengths: polaron states (1), (2) and (3).

In the absence of LO-phonon coupling, the only optically active excitonic states in the low energy absorption spectrum of a QD, are those in which both the electron and the hole are in p-states. However, when including Frohlich coupling, LO-phonon replica of additional states needs also to be taken into account. This leads to a more complex absorption spectrum. In particular, with increasing magnetic field, a crossing is predicted between a bright excitonic state (with both electron and hole in the p-states) and a LO-phonon replica of a dark excitonic state (with the electron in the s state and the hole in the p state). When LO-coupling is taken into account, the crossing of these two states leads to the formation of the two polarons states labeled (1) and (2) in Figure 5.

Figure 6 shows a comparison between the calculated absorption peaks and the observed RPL for E_{exc} =

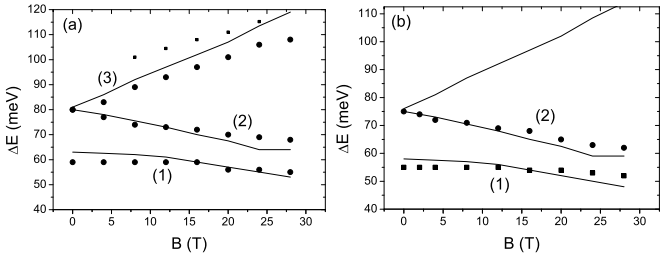

Fig. 6. Magnetic field dispersion of RPL resonances for E_{exc} = 1293 meV (a) and E_{exc} = 1260 meV (b) in symbols. The solid lines are the calculated polaron energy levels.

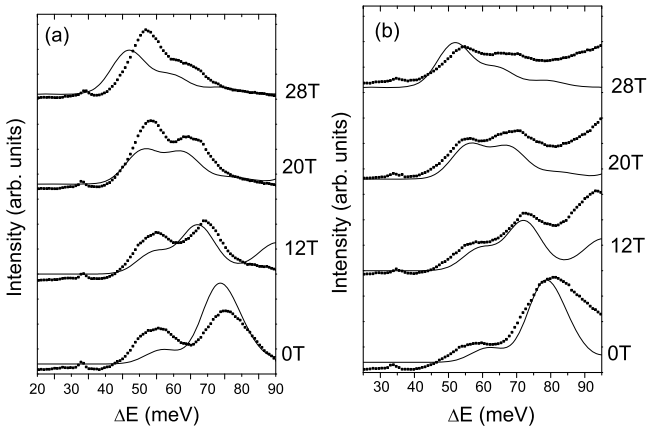

Fig. 7. Experimental (full symbols) and calculated (solid lines) spectra for different magnetic fields. The experimental data was taken for E_{exc} = 1260 meV (a) and E_{exc} = 1293 meV (b), for sample P3. The parameters used in the calculation are given in the text.

1293 meV (a) and E_{exc} = 1260 meV (b). The calculated energy position as a function of magnetic field are found in good agreement with our data. Polaron level (3) is not experimentally observed for E_{exc} = 1260 meV for reasons explained in the previous section. In addition we note that, as mentioned in the previous section, the detection energies of the peaks in the E_{exc} = 1293 meV spectra are different from the detection energies of the peaks in the E_{exc} = 1260 meV spectra. This in turn means that different sized dots were selected for the two different excitation energies. In order to account for this, we have performed a rigid upshift on the calculated polaron levels in order to fit the experimental results. The calculation was shifted up by 10 meV in Figure 6a and by 5 meV in Figure 6b. By performing the necessary shift, we are able to use the same dot parameters for measurements that select different sized dots. This accounts for the difference in energies of the calculated polaron states in Figure 6a as compared to Figure 6b.

The solid lines in Figure 7 represent the calculated polaron absorption spectra at different magnetic fields, again taken from reference [2]. Each discrete polaron level was replaced by a Gaussian peak with a *FWHM* of 12 meV in order to account for inhomogeneous broadening (i.e. fluctuations of dot dimensions in the subensemble of QDs defined by the excitation energy). The symbols in Figure 7

are data points taken for the p-doped sample for $E_{exc} =$ 1260 meV (a) and $E_{exc} = 1293$ meV (b).

We remark slight differences in the experimental spectra of Figure 7a as compared to the calculations. Firstly, at 0 T, we experimentally observe very similar intensities for the two peaks, whereas the calculated spectrum predicts a stronger oscillator strength (OS) for the high energy branch. In addition, at 20 T, where we predict equal OS's for both peaks, the experimental data displays a lower energy peak more intense than its high energy neighbor. The tendency for the high energy peak to be weaker than what is theoretically predicted can be explained using the reasoning presented in the previous section. Due to the dot size inhomogeneity present in our samples, only a small population of dots will be found at high energies for this low excitation energy. Our calculated spectra were found using one particular dot size and therefore do not take into account any size inhomogeneity. This could therefore be an explanation of the slight discrepancy observed between the experimental and theoretical results in Figure 7a.

On the other hand, the spectra of Figure 7b was measured at an excitation energy, $E_{exc} = 1293$ meV. There exists a large population of dots with excited states equal to this excitation energy. The difference in intensity of the two observed peaks, can therefore be attributed fully to a polaron anti-crossing effect and not the dot population distribution. We find here that the evolution of the peak intensities with the magnetic field is very well described by our model. We are able to predict the exchange of oscillator strength observed in both our results, demonstrating the validity of our analysis. The unequivocal anti-crossing observed in the RPL spectra provides evidence of the existence of excitonic polaron states and confirms previous PLE measurements on similar samples [2].

Finally, up until now we have not attempted to explain the high energy peak at ∼110 meV, observed in our experimental results (Fig. 1). This high energy peak, that has also been detected in previous PLE measurements [2], stays at a nearly constant energy as the magnetic field is increased. Our excitonic polaron calculation does not predict such a peak with a strong oscillator strength in this energy range. The evolution of this peak in a magnetic field is reminiscent of the behavior of transitions that involve s-like states, whose energy moves very little (diamagnetic effect) with an applied B field. In addition, when calculated, the optically allowed transition involving excited states of s symmetry ($2s_e$–$2s_{hh}$) is found to be ∼160 meV above the ground state excitonic level, and therefore too high to account for this peak.

The QD energy level calculation used here does not include the light hole states. We assume, due to the strong strain in a self-assembled QD system, that these states are very close in energy to the GaAs valence band edge [15] and therefore will not be pertinent to the description of our experimental results.

However, recent theoretical studies by Narvaez and Zunger [11] have revealed the importance of the light hole states. Using an atomistic pseudopotential approach [16], the strain-modified valence band offset of dots similar to those of our samples were calculated [11]. A confined light hole state was found below that of the heavy hole. In addition, the excitonic absorption spectra of the QDs were calculated where a significant peak approximatively 100 meV above the ground state energy is predicted. The peak corresponds to an interband transition between the ground conduction band state of s symmetry ($1s_e$) and the ground light hole state of s symmetry ($1s_{lh}$). This transition is a possible explanation for the high energy peak observed in our experimental results. However, a definitive identification of this transition is difficult because experiments were performed with unpolarized light and involve an ensemble of dots whose shape and orientation vary from dot-to-dot. Moreover, the PL intensity cannot be used as a discriminating criterium because light and heavy hole relaxations are not expected to be equivalent. Performing single dot spectroscopy would alleviate these difficulties. In addition, performing single dot spectroscopy using polarized light would certainly help to clarify the status of this transition.

5 Conclusion

In summary, we have investigated the interband transitions in several ensembles of self-assembled InAs/GaAs QDs by using RPL spectroscopy under strong magnetic field. The RPL technique allows us to overcome the homogenous broadening of the transitions because only subensembles of similar QDs are excited and contribute to the PL signal. Moreover, contrary to PLE spectra, the RPL spectra does not include the large peak of the wetting layer. The observed well defined peaks are found in good agreement with previous calculations that include exciton LO-phonon couplings. In particular, transitions involving excitonic polarons are clearly identified by the anticrossing occurring when two exciton-LO phonon states have close enough energies with phonon occupations which differ by one. Moreover, a high energy peak was observed that may be attributed to a light hole to conduction transition in agreement with recent calculations based on an atomistic pseudopotential approach [11].

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