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"Evolution of the optical properties of InAs/GaAs quantum dots for increasing InAs coverages"

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Evolution of the optical properties of InAs/GaAs quantum dots for increasing InAs coverages

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Abstract

The photoluminescence (PL) of InAs/GaAs heterostructures is investigated for InAs coverages, L, ranging from 0.6 to 3 monolayers (ML). For thin coverages ($L \leq 1.6$ ML), we observe the recombination of heavy-hole excitons in InAs quantum dots (QD's) and in a 2D-InAs layer. The two PL bands shift toward low energy for increasing L. For $L \geq 1.6$ ML, the QD band shifts faster, while the exciton recombination in the 2D-layer vanishes. These results, confirmed by PL excitation and photoreflectivity, indicate that: a) QD's are interconnected by a two-dimensional InAs layer which allows an efficient carrier capture into the dots; b) the dot size increases with L, faster for $L \geq 1.6$ ML, at the expenses of the 2D-layer. The peculiar temperature dependence of line shape and peak energy of the QD band is explained in terms of exciton thermal escape and relaxation mechanisms.

Subject classification: 71.55.Eq; 78.55.Cr; 73.20.Dx; 79.60.Jv.

In recent years, a strong effort has been devoted to the study of highly strained InAs/GaAs heterostructures grown by molecular beam epitaxy (MBE). The stress present at the interface between InAs and GaAs provides a natural driving force for the self aggregation of InAs islands, which behave like quantum dots, QD's. These nano-structures open new outlooks for the realization of modern optoelectronic devices, such as low threshold injection lasers and single electron devices. [1] Because of the strong dependence of the MBE growth on kinetic factors, it is difficult to obtain the strict control of the dot morphology required for the realization of devices. Therefore, a detailed study of the growth is necessary. In spite of the large number of experimental and theoretical papers on this topic, only a few of them deal with the evolution of the InAs layer morphology with InAs coverage, L. [2,3] Previously, [2] our group has shown that QD's nucleate for $L < 1.6$ ML, the critical thickness commonly reported in literature. [4]

In this work we investigate the optical properties of InAs/GaAs heterostructures for $0.6 \leq L \leq 3$ monolayers (ML), by photoluminescence (PL), PL excitation (PLE), and photoreflectivity (PR). Our measurements show that QD's are interconnected by a 2D-InAs layer, at least for thin coverages. The dot size increases faster for $L \geq 1.6$ ML, at expenses of the 2D layer. Besides, a structured QD-PL-band suggests a multimodal distribution of the QD size. Finally a peculiar dependence on temperature of line shape and peak energy of the QD band is explained in terms of an exciton thermal escape and relaxation mechanisms.

Two sets of GaAs/InAs/GaAs heterostructures have been grown by MBE on a GaAs (001) substrate, in two different chambers, at two temperatures (T = 420 °C for $0.6 \leq L \leq 2$ ML, and T = 500 °C for L = 1.8, 3 ML). PL and PLE measurements have been performed, at different T, by using a Ti:Sapphire and a Ar^+ laser. PR was performed at room temperature by using a halogen lamp as probe source and a He-Ne laser as excitation source.

The PL, PLE and PR spectra for samples with L equal to 1.2 and 2 ML are shown in Fig. 1. In order to compare PR at 295 K with PL and PLE at 5 K, PR has been translated to high energy by 90 meV, i.e., the measured thermal energy shift of the GaAs bandgap.

In the 1.2 ML sample, the PL shows two emission bands: HH and QD. The former is due to the heavy hole free exciton recombination in a 2D-InAs layer of 1.2 ML, the latter originates from an exciton recombination in InAs QD's. The HH resonance is also observed in the PLE and PR spectra with a Stokes shift of 6 meV. Moreover, the light hole free exciton resonance in the 2D-layer (LH) is revealed both in PLE and PR spectra. Finally a weak absorption from the dots is revealed only in the PLE spectrum. The nature of the HH and LH resonances is excitonic up to 295 K, as confirmed by a best fit to the room temperature PR spectra. In fact, two excitonic line shape functional form models, typical of quantum confined systems, [5] have to be used to reproduce the experimental PR data. Similar results are found for all samples with L between 0.6 and 1.6 ML.

For $L > 1.6$ ML, PL shows a broad QD band, with a full width at half maximum (FWHM) of about 40 meV, while the HH and the LH resonances are not observed in PL as well as in PLE and PR. As shown in Fig. 1, the QD band is observed in the PLE spectrum with a Stokes shift of about 20 meV for the 2 ML sample.

It has to be stressed the absence of QD related features in the PR spectra of all samples.

The HH and LH energies, measured by PLE and PR, are shown in Fig. 2(a) and compared with calculations based on a quantum-well model where the strain and the Coulomb interaction are taken into account. We assume a band offset equal to 0.7, a well width equal to the nominal InAs coverage, and an exciton binding energy of the HH and LH (4.2 and 2.8 meV, respectively) equal to those obtained for the GaAs free exciton in a 3D-hydrogenic model. As shown in figure, the transition energies measured by the peak positions in PR coincide with those obtained from PLE. The good agreement with the model supports the assignement of the HH and LH emissions to the 2D-InAs layer.

The peak energy of the QD PL band at $T = 5$ K is reported as a function of L in Fig. 2(b). The QD band shifts toward low energy for increasing L, faster for $L \geq 1.6$ ML, where it becomes structured, as shown by the two QD-peak-energies reported for the 3 ML sample.

PLE spectra, which are related to both the generation and relaxation of carriers, provide evidence of an interconnection between the QD's and the 2D-InAs layer, at least for thin coverages. In fact, the HH and LH resonances are observed when monitoring the PL at the QD states, thus showing that excitons created in the 2D-layer are efficiently captured into QD's where they recombine. This accounts for the high QD PL efficiency, despite the prediction of a phonon bottleneck for carrier relaxation. [6] In this case, indeed, the 2D-layer provides a continuum of states which favors the carrier thermalization into dot states.

The lack of a 2D related optical absorption in PR and PLE spectra, and the faster shift of the PL QD band at large L, suggest a change in the self aggregation regime: the dot size increases faster for $L \geq 1.6$ ML, at the expenses of the 2D-InAs layer, which shrinks. At the same time the appearance of a structured QD-PL-band suggests a multimodal distribution of the dot size. For what concerns the PR spectra, the absence of features related to QD's may be due to the destructive interference of the randomly-phased optical responses of a large number of different single dots or to a lack of sensitivity.

A QD multimodal distribution may account for the peculiar shift with increasing temperature of the QD-PL-peak energy, measured with respect to the value at T = 10 K and reported in Fig. 3. Therein, the shift for samples with L equal to 1.2, 2 and 3 ML is compared with the shift of the GaAs free exciton, FE. In the 1.2 ML sample, the QD peak energy closely follows that of the GaAs FE; on the contrary, in the 2 and 3 ML samples, the QD band shifts faster toward low energy for $T > 70$ K and moves back toward the GaAs FE curve at higher T. This behaviour may be explained in terms of thermalization processes and thermal escape processes from QD's. In fact, QD's with smaller sizes and high energy states are depleted for increasing T, thus causing a faster thermal red shift. This effect may be due also to a partial thermalization of carriers within dots by tunneling processes and carrier diffusion through a residual 2D-InAs layer. At higher T, the exciton population of high energy QD's should be re-established to account for the behaviour reported in Fig. 3 for $T >$ 130 K. This peculiar behaviour with T is not observed in the 1.2 ML sample due to the small distribution in the QD size suggested by a narrow PL-QD-band ($\approx$ 20 meV). The carrier thermalization within dots is supported by the large QD absorption revealed in PLE by monitoring the luminescence on a single QD level and by the large Stokes shift of the QD band for the 2 ML sample (see Fig. 1).

The temperature dependence of the FWHM of the QD band, reported in the inset of Fig. 3 for the 3 ML sample, is fully consistent with the above analysis. The difference, DE, between the GaAs FE-band and the QD band increases whenever the FWHM decreases and *viceversa*. The processes invoked to explain the red shift of the QD band should result also into the QD band narrowing observed at intermediary T, while the re-establishment of the QD-exciton population should give rise to the broadening reported at high T.

In conclusion, a detailed optical analysis of a set of InAs/GaAs heterostructures grown by MBE has shown that InAs-self-aggregated QD's are interconnected by a 2D layer, at least for thin L. For $L > 1.6$ ML, the 2D layer shrinks in favor of QD's. Moreover, a multimodal QD distribution accounts for a peculiar temperature dependence of the PL-QD-band, which is explained in terms of an exciton thermal escape and relaxation processes.

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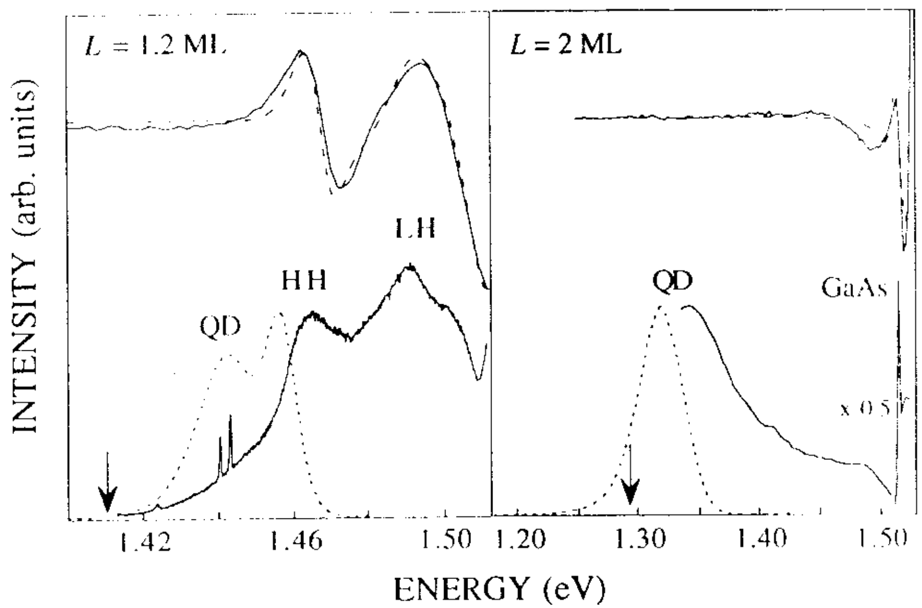
Figure Captions

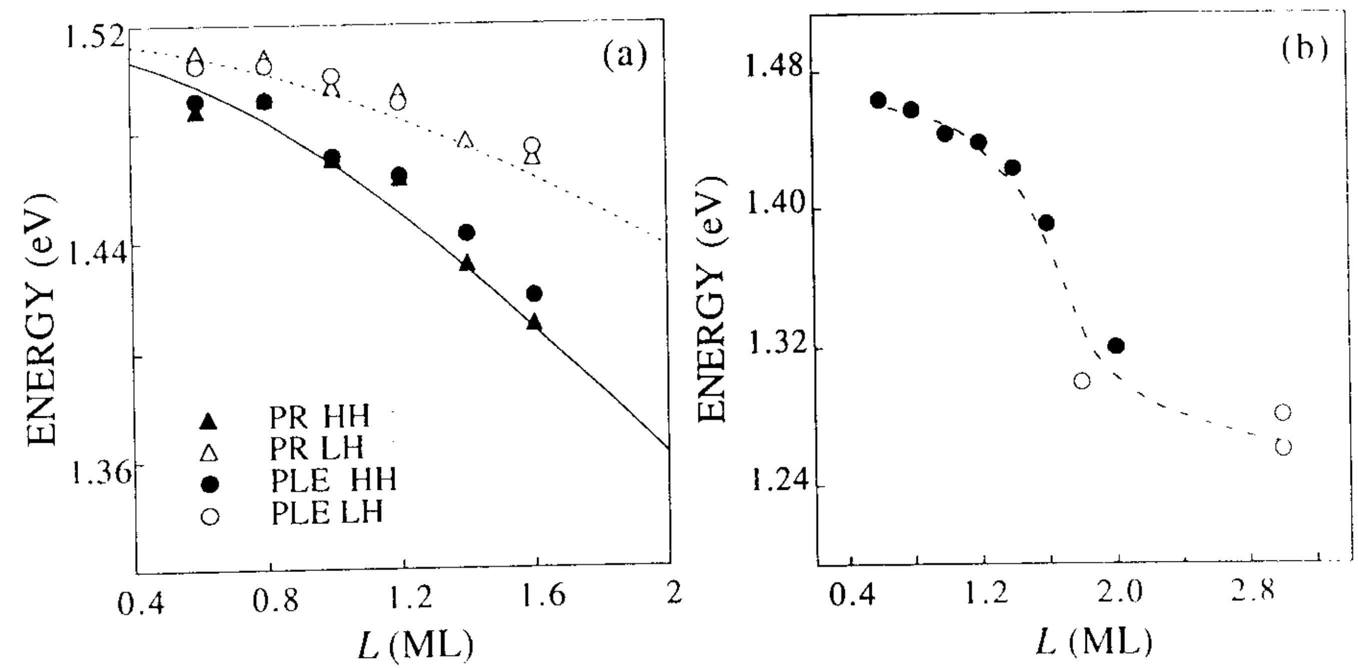
Fig. 1: PL (dotted line), PLE (full line at the bottom) and PR (full line at the top) spectra for the samples with L equal to 1.2 and 2 ML. The dashed line is the best fit to the PR data. The detection energies in the PLE spectra are 1.41 eV and 1.29 eV for the 1.2 ML and 2 ML samples, respectively (see arrows).

Fig. 2: (a) Energies of the HH (full symbols) and LH (open symbols) measured by PLE (dots) and PR (triangles). To compare PR at 295 K with PLE at 5 K, PR has been shifted to high energy by 90 meV. The lines give the exciton energy as calculated in a quantum well model. (b) QD PL peak energy at 5K as a function of L. Full and open dots refer to samples grown at 420 °C and 500 °C respectively. The dashed line is a guide to the eye.

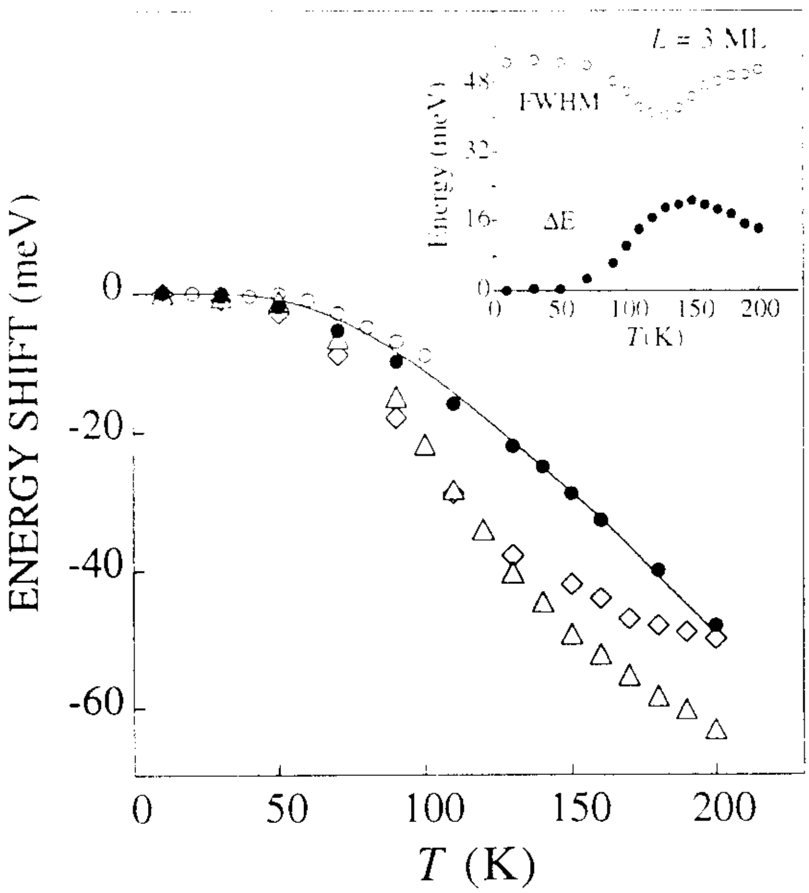
Fig. 3: Energy shift of the QD PL band, measured with respect to the value at T = 10 K. Open dots refer to L = 1.2 ML, diamonds to L = 2 ML, triangles to L = 3 ML. The shift of the GaAs FE (full dots) and the temperature dependence of the GaAs bandgap (full line) are also reported. In the inset, the temperature dependence of the FWHM for L = 3 ML is reported, together with the energy difference, DE, between the GaAs-FE band and the QD band.



A. Patanè et al.: Fig. 1



A. Patanè et al.: Fig. 2



A. Patanè et al.: Fig. 3

OPTICAL PROPERTIES OF InAs/GaAs QUANTUM DOTS FOR INCREASING InAs COVERAGES

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OUTLINE

HIGHLY STRAINED InAs/GaAs HETEROSTRUCTURES

Absence of a critical thickness for the 2D-3D transition

Two growth regimes for increasing InAs coverages

Electronic states and relaxation mechanisms in QD's

EXPERIMENT

Samples

InAs/GaAs heterostructures grown by MBE

InAs coverage, L, between 0.6 and 3 ML

Optical techniques:

Photoluminescence (PL)
PL excitation (PLE)
Photoreflectance (PR)

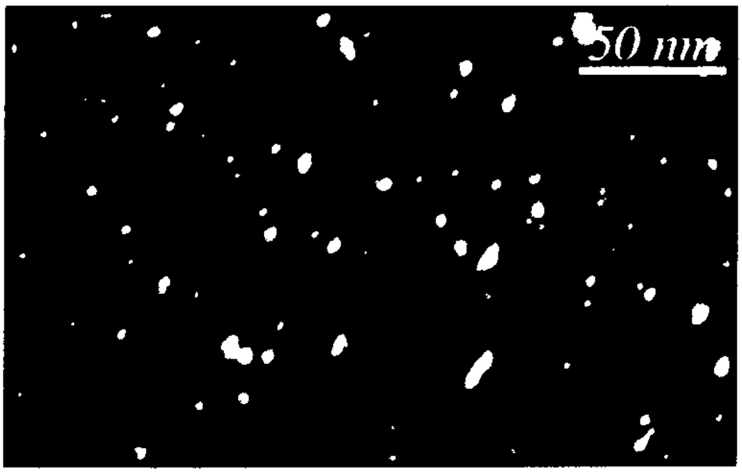
Microscopical technique:

Transmission Electron Microscopy (TEM)

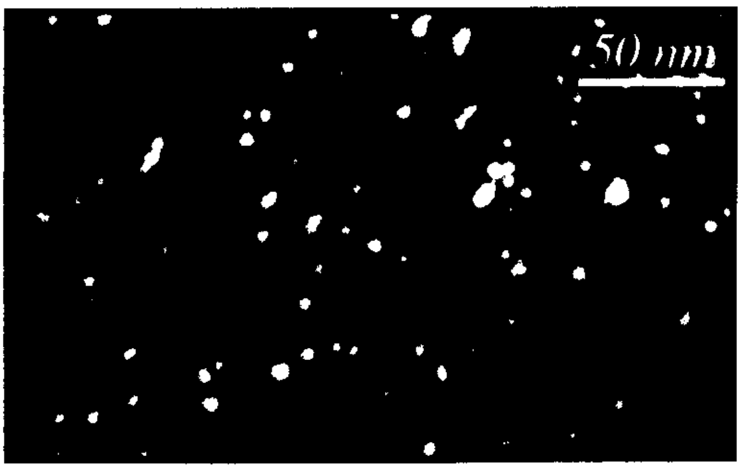
PLANAR TEM

G. Salviati et al, *CNR-MASPEC (Parma)*

$L = 0.8$ ML



$L = 1.2$ ML

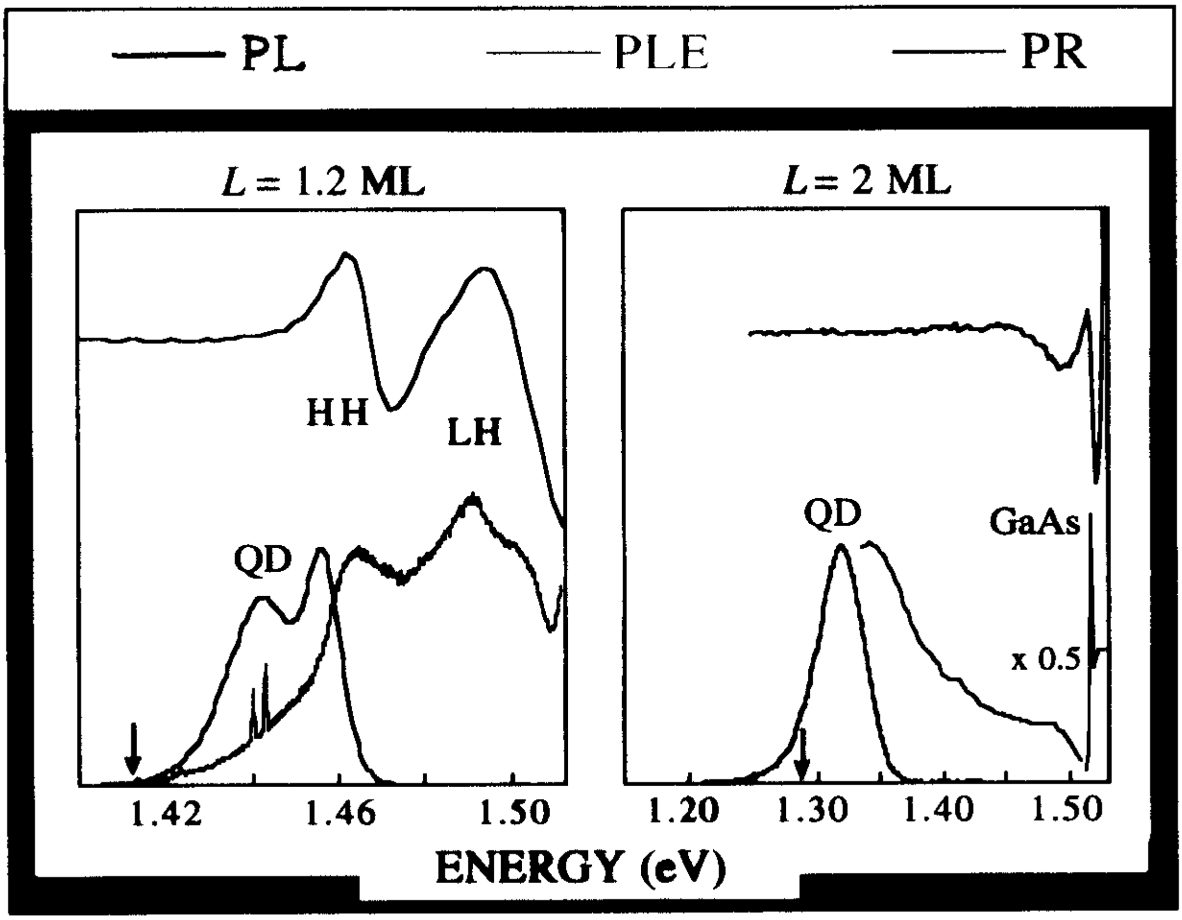


QD formation for thin L

TRANSVERSE TEM

G. Salviati et al, *CNR-MASPEC (Parma)*

2D AND 0D STATES



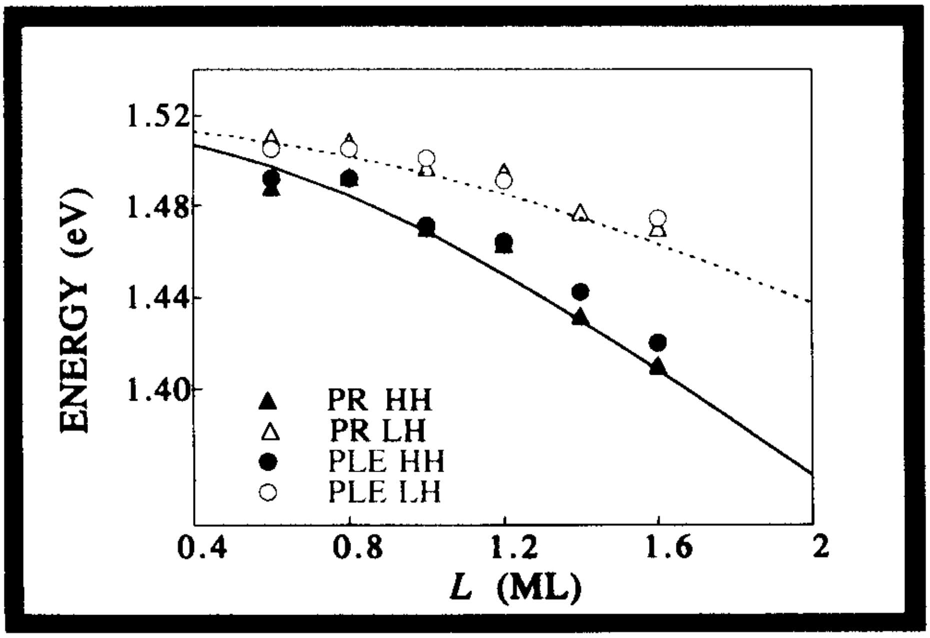
$L \leq 1.6$ ML → QD's and the 2D layer

$L > 1.6$ ML → Lack of the 2D layer

THE 2D-InAs LAYER

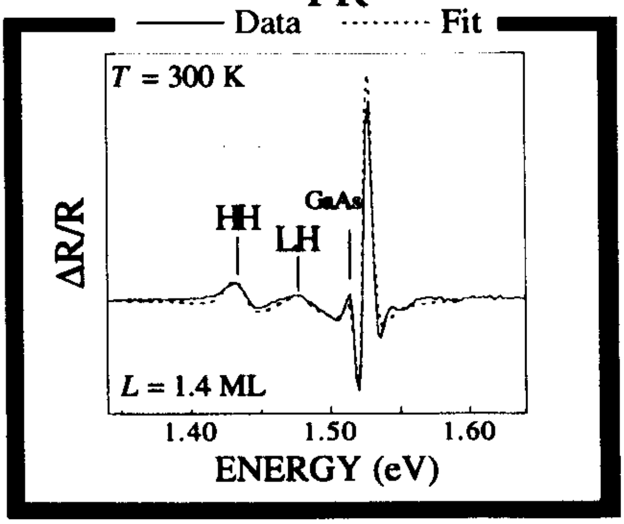
A square QW model for the 2D-InAs layer

HH:—— LH: -----

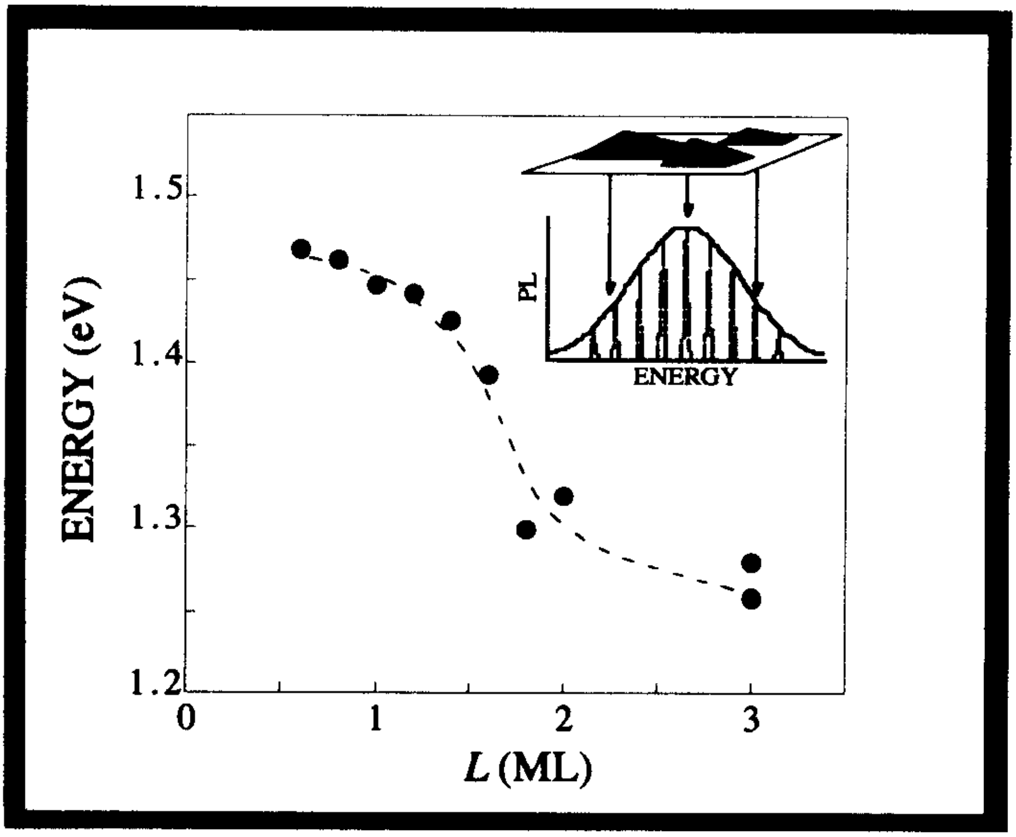


Excitonic origin of the HH and LH peaks up to 300 K

PR



SELF-AGGREGATED QD'S



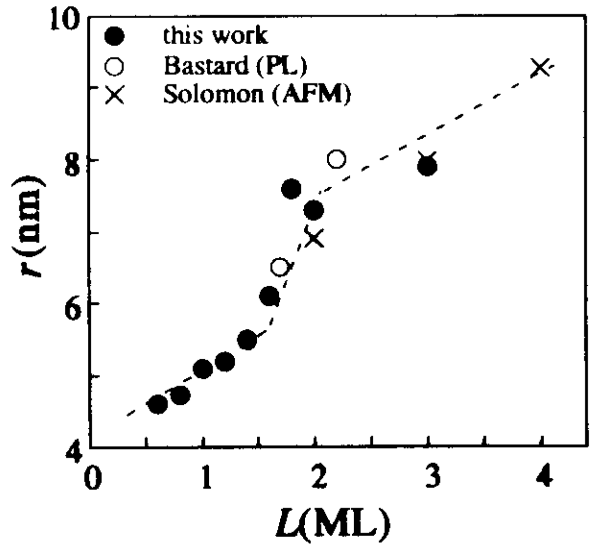
By increasing *L*:

- the dot size increases
- the 2D-InAs layer shrinks
- the QD band becomes structured

QD Morphology

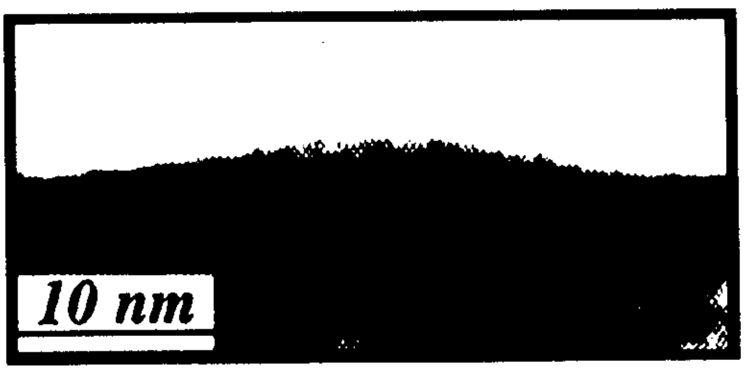
QD size vs L

Model for QD's
J.-Y. Marzin and G. Bastard
Solid State Comm. 92 (1994)



High resolution TEM

$L = 3$ ML



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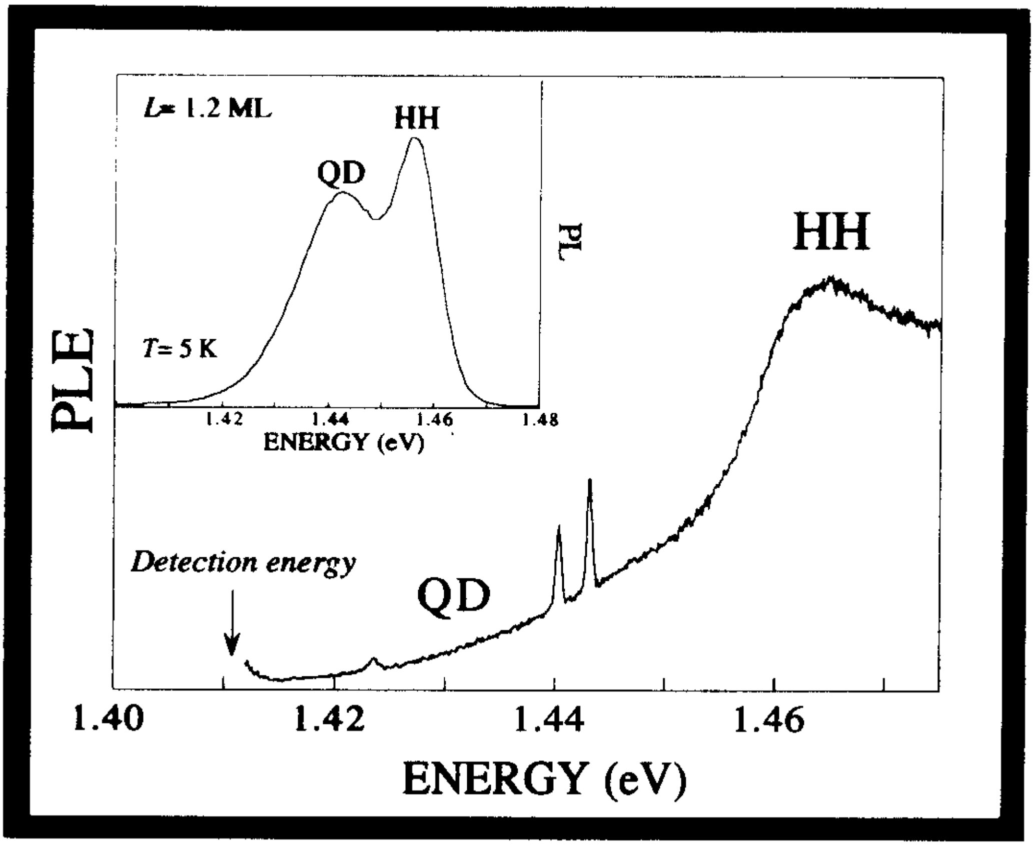
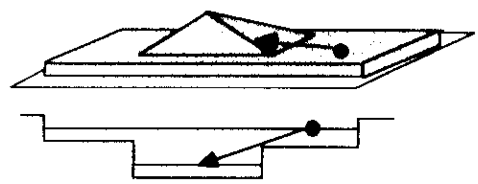
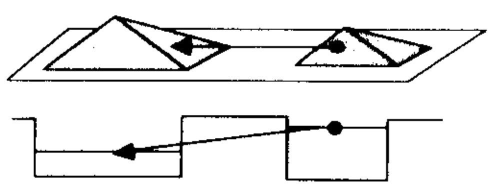
CARRIER RELAXATION



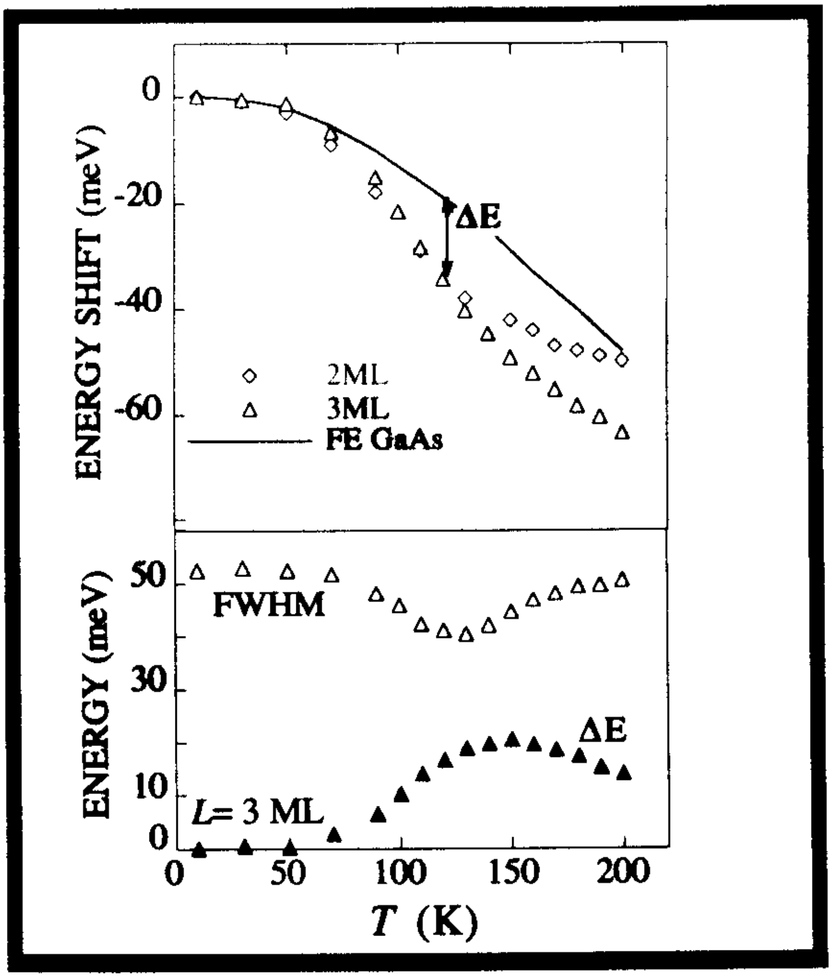
Diffusion through the 2D-layer



Tunneling between QD's



ANOMALOUS THERMAL EFFECTS

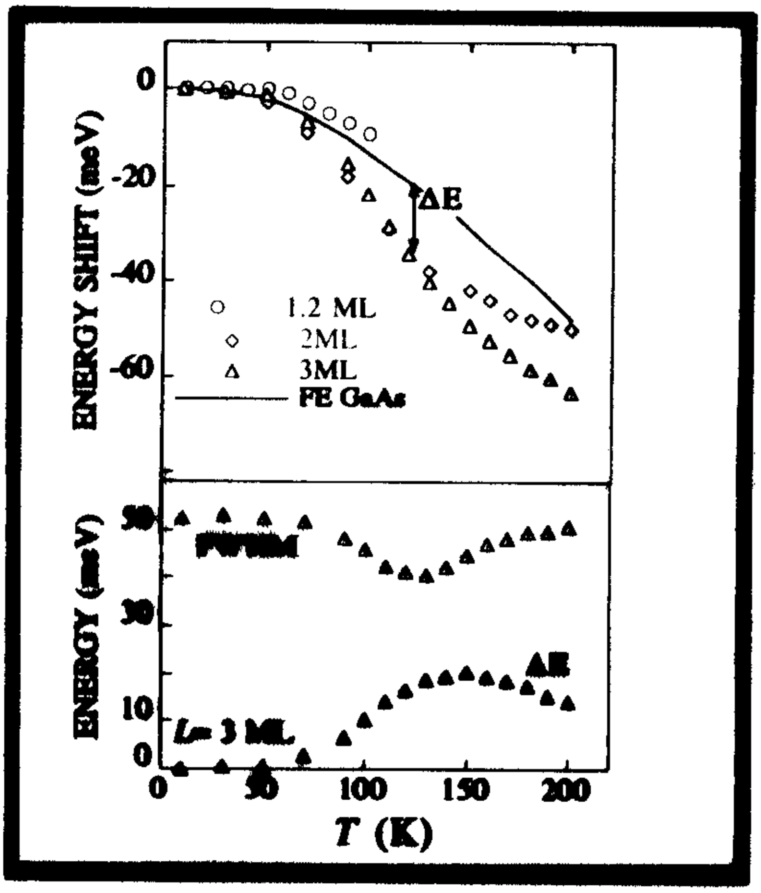


The red shift is due to:

Carrier *thermal escape from* dots

Carrier *thermalization between* dots by
- diffusion through a residual 2D layer
- tunneling processes

Anomalous Thermal Effects



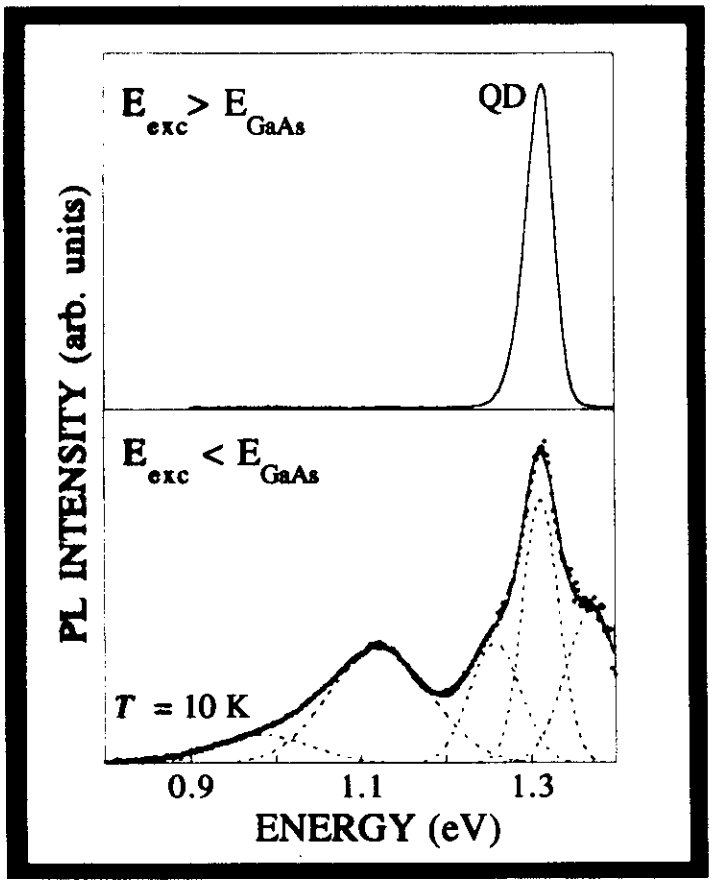
The red shift is due to:

Carrier *thermal escape from* dots

Carrier *thermalization between* dots by
- diffusion through a residual 2D layer
- tunneling processes

QD Families or InAs-Related Defects?

$L = 2$ ML



Excitation below the GaAs gap ⟶ additional PL bands

CONCLUSIONS

Continuous Self-Aggregation of QD's

Two Growth Regimes

Thin L → QD's and the 2D layer

Thick L → predominant growth of QD's

Carrier Thermalization Between QD's

Diffusion through the 2D layer

Tunneling processes