Optical spectroscopy of quasimonolayer InAs at the onset of quantum-dot nucleation

A. S. Bhatti,* M. Grassi Alessi, and M. Capizzi
Istituto Nazionale di Fisica della Materia, Dipartimento di Fisica, Università di Roma ''La Sapienza,'' Piazzale A. Moro 2, I-00185 Roma, Italy

P. Frigeri and S. Franchi
CNR–MASPEC Institute, Parco Area delle Scienze, I-43100 Parma, Italy
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We have performed low-temperature photoluminescence (PL), resonant PL (RPL), and PL excitation (PLE) measurements on different series of self-organized InAs/GaAs quantum dots (QD's) in samples with an InAs nominal coverage (L) varying from 1.2 to 3 monolayers (ML). Drastic changes in the PL spectra have been observed for L values spanning across the so-called critical thickness L_c ($\sim$1.7 ML). RPL has shown that both QD's and quasi-three-dimensional QD precursors contribute to the spectra of samples with $L \lesssim L_c$. PLE measurements have allowed us to introduce an accurate determination of L_c and to verify its dependence on sample growth conditions. Finally, the analysis of PL spectra in all investigated samples and its comparison with spatially resolved PL measurements has suggested a different interpretation of doublet and triplet bands usually found for $L \sim L_c$ and previously ascribed to excited states or multimodal distributions of QD families. [S0163-1829(99)05627-1]

I. INTRODUCTION

The search for zero-dimensional structures either for application purposes or for fundamental studies is an intensely pursued research topic.[1] In particular, the prediction of improved laser device performances at room temperature has given strong support to research on quantum dots (QD's).[2] Although growth mechanisms and optical properties have been widely investigated and greatly improved, a number of issues are still a matter of debate. These are, for instance, the dependence on growth conditions of the critical thickness (L_c) for the self-aggregation and evolution of QD's, the effects of In and Ga interdiffusion for different coverages, etc. All these features strongly affect the optical properties of the dots and the overall device performance.

Recently, we have shown that QD's in InAs/GaAs heterostructures with the same InAs nominal coverage (L) emit at a number of quantized, discrete energies.[3] This has been interpreted in terms of different QD shapes and of their dependence on growth conditions. These results hold also for other lattice mismatched systems, e.g., to $InP/Ga_xIn_{1-x}P$ heterostructures.[4] According to Ref. 3, the evolution of QD's with InAs coverage defines three distinct L regions, I–III, as shown in Fig. 1 where the emission energy of the strongest QD band (E_p) is reported as a function of L. Solid symbols refer to samples further investigated in the present work, open symbols refer to samples previously prepared in a different laboratory under different growth conditions.[5] The second, intermediate region (II), spans across the onset for the formation of quantum dots and exhibits the most drastic dependence of the optical properties on L. In this region, the QD emission energy falls off quickly with L. Moreover, QD photoluminescence (PL) bands are characterized by doublets or triplets with roughly constant energy separation ($\sim$30 meV), as it will be shown in the following, or by singlets broader than those of samples in regions I or III. Finally, PL bands related to the recombination of heavy-hole (HH) and light-hole (LH) excitons in the two-dimensional (2D) layer underlying the dots, usually called wetting layer (WL), dominate the PL excitation (PLE) spectra.

In the present paper, we discuss the most striking aspects of the above-mentioned intermediate growth region. A smooth transition takes place from region I, characterized by structures that can be identified as QD precursors, to region II, where both precursors and QD's coexist. The QD precursors are capable of confining carriers and giving rise to PL bands below the WL energy. Those bands can be distinguished from the bands due to fully formed QD's by performing PL experiments in a crossed excitation/detection geometry. The same type of multiplets with roughly constant energy separation ($\sim$30 meV) observed in the PL bands of

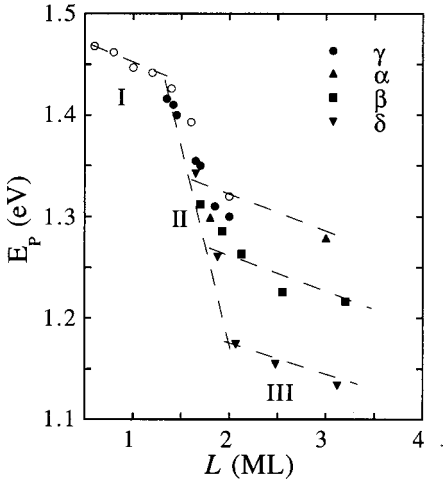

FIG. 1. Peak emission energies vs L of the main bands observed in the PL spectra of all the samples listed in Table I. Dashed lines are guides to the eye and define three different regions in the plane, I–III. Data from Ref. 5 are reported as open circles.

TABLE I. Specifications of the samples investigated in the present work. The relative uncertainty on sample thickness is ±5%. All samples have a GaAs cap layer of 20 nm.

Samples	L (ML)	T_G (°C)
α	1.8, 3.0	500
β	1.5, 1.7, 1.9, 2.1, 2,6, 3.2	500
γ [a]	1.2, 1.25, 1.35, 1.4, 1.45	460
γ	1.65, 1.7, 1.8, 2.0	460
δ	1.2, 1.45, 1.7, 1.9, 2.1, 2.5, 3.1	520

[a]The samples have been prepared in a single growth on a nonrotating holder.

samples in region II are present also in the PL bands of samples with L quite different from L_c. These features and PL measurements as a function of exciting power density allow us to exclude that those multiplets can be explained in terms of excited states or different QD families. We tentatively explain those multiplets in terms of carrier interaction with InAs longitudinal-optical phonons. Finally, we study the 2D to 3D transition and the onset for the aggregation of fully developed QD's. Theoretical models generally predict that the InAs/GaAs system, under thermal equilibrium conditions, follows the Stranski-Krastanow growth mode. In particular, QD's are expected to nucleate spontaneously over an InAs layer only when a critical thickness of InAs has been deposited. This thickness has been identified with a nominal coverage of about 1.7 ML by reflection high-energy electron diffraction (RHEED) and microscopical measurements. However, recent theoretical calculations[6,7] have shown that the critical thickness value should depend on sample growth conditions and on the sensitivity of the experimental technique used for its determination. In order to support these theoretical results, we have introduced an empirical determination of L_c by means of PLE experiments whose sensitivity to the formation of QD's is higher than that of RHEED. We show that the critical thickness value depends on growth temperature. Moreover, the WL growth is subject to fluctuations until it is frozen in a disordered morphology by the overcoming nucleation of dots. These results indicate that strong kinetic effects, usually not included in the theoretical models, partially mask the thermodynamic equilibrium conditions during the QD growth.

II. EXPERIMENT

Low-temperature photoluminescence, resonant PL (RPL), spatially resolved PL (μPL), and PLE spectroscopy were performed on InAs/GaAs samples grown by molecular-beam epitaxy under different conditions. Sample specifications have been summarized in Table I. Optical measurements were performed at 10 K, unless otherwise specified, in a closed cycle He optical cryostat by means of a He-Ne or an ion Ar^+ laser and of a Ti-sapphire tunable laser. The signal was spectrally analyzed by a double monochromator and collected by a photomultiplier or an $In_xGa_{1-x}As$ detector. All the spectra were corrected in order to take into account the response of the system. The PL and PLE experiments were carried out at various laser power densities with a laser spot size variable between 1 and 200 μm.

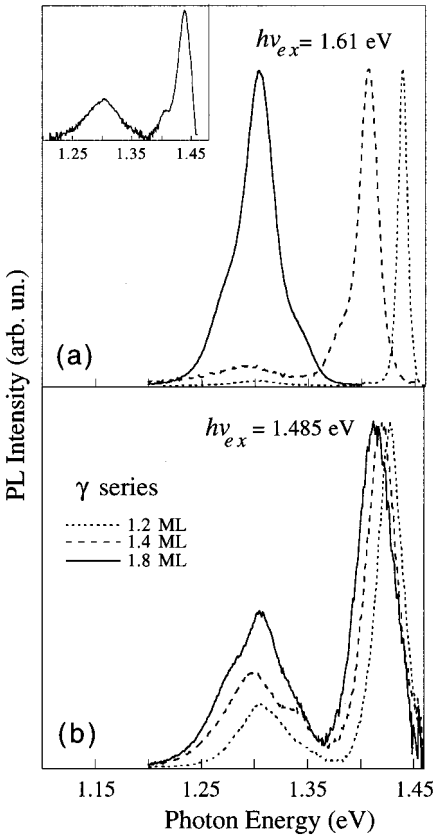

FIG. 2. PL spectra of three samples from γ series. The spectra are taken at 10 K with excitation energy (a) above, 1.61 eV, and (b) below, 1.49 eV, the GaAs band gap, 1.519 eV. The excitation density is 1 Wcm^{-2}.

III. RESULTS AND DISCUSSION

In Fig. 2, we show the PL spectra of three samples of the γ series with L between 1.2 and 1.8 monolayer (ML) for excitation energies (E_{ex}) above [E_{ex}=1.61 eV, nonresonant excitation, Fig. 2(a)] and below [E_{ex}=1.485 eV, resonant excitation, Fig. 2(b)] the GaAs band gap. The nonresonant PL spectrum of the L=1.2-ML sample exhibits a sharp peak at 1.44 eV, which is due to the recombination of excitons confined in the 2D InAs layer. By a careful analysis of the spectrum, two additional bands, broader and one hundred times weaker than that at 1.44 eV, are identified at about 1.41 and 1.3 eV; see the inset in the figure where the logarithm of the PL intensity is reported. By increasing the InAs coverage to 1.4 ML, the band related to the 2D layer disappears and the 1.41 eV broadband becomes dominant, while the 1.3 eV band is still weak. After a further increase of the InAs coverage to L=1.8 ML, the relative weight of the two broadbands is reversed.

The PL spectra taken for resonant excitation in the same three samples reported in Fig. 2(a) are shown in Fig. 2(b). These spectra exhibit two main bands of comparable weight, separated by more than 100 meV and roughly coincident with the two broadbands observed in the case of nonresonant excitation. The simultaneous presence of both bands for all coverages in the γ series, with comparable PL intensities, is confirmed by RPL measurements. We ascribe these two bands to the recombination of carriers confined in two dif-

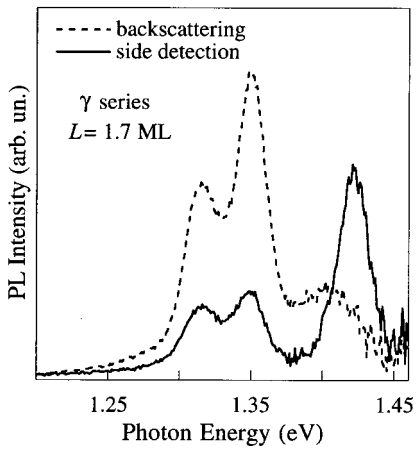

FIG. 3. PL spectra of the 1.7-ML γ sample detected along a direction parallel (solid line) or perpendicular (dashed line) to the InAs 2D layer. In both cases, the excitation is perpendicular to the 2D InAs layer.

ferent zero-dimensional structures, i.e., two different types of QD's. Under resonant conditions, carriers are expected to be directly and selectively generated in a reduced number of QD excited states and, thus, to recombine only in the corresponding dots. On the contrary, for non-resonant excitation, free carriers are mainly generated in the GaAs barrier. Thus, the resulting PL line shape depends on the complex mechanism of carrier capture by the QD, eventually leading to different probability of relaxation into different types of dots.

Further measurements under resonant conditions clarify the different nature of the two types of QD's. Two PL spectra detected in the 1.7-ML sample of the γ series for different excitation/detection configurations are shown in Fig. 3. In the standard backscattering geometry—both excitation and detection along the direction perpendicular to the InAs layer—a doublet appears at energies below 1.35 eV, while a weaker, broad structure is observed at $\sim$1.40 eV. When detecting the PL along the InAs plane—the excitation being maintained along the direction perpendicular to the InAs layer—a peak at 1.42 eV dominates the PL spectrum. We ascribe the two bands emitting at lower energies and insensitive to the detection geometry to ordinary, three-dimensional QD's shaped like pyramids, an assignment confirmed by transmission electron microscopy (TEM) measurements.[8] We ascribe, instead, the 1.42 eV band, which lies at an energy lower than but close to that expected for a 2D layer, to recombination of carriers confined in structures with a lens-shaped geometry and low aspect (height to base) ratio. In fact, PL bands with features similar to those of the 1.42 eV band have been observed previously in a series of samples grown in a different laboratory that are characterized by the presence of lens-shaped structures, as shown by high resolution TEM measurements.[5,9] In these structures, the dependence of the PL peak energy on L practically coincides, as shown by the open symbols in Fig. 1, with that of the higher-energy band at $\sim$1.42 eV reported here for the samples of the γ series. We assume that such flat structures with almost 2D geometry are morphologically linked to the disordered 2D InAs layer. In that case, a sizable waveguiding effect can lead to a strong increase of PL intensity for detection along the plane of the 2D InAs layer, as observed here.

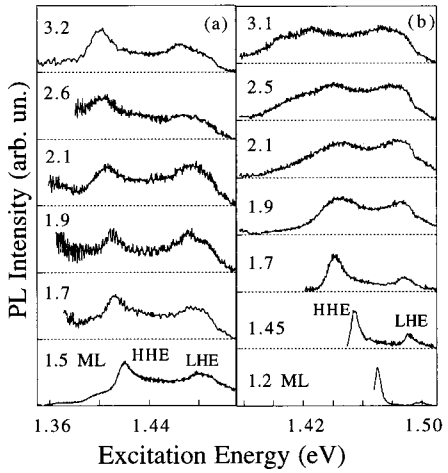

FIG. 4. PLE spectra for (a) the β and (b) the δ samples. The heavy-hole and light-hole excitons in the wetting layer (HHE and LHE, respectively) get broader and shift to lower energies for increasing L. Multiple structures are observed in the PLE of δ samples for $L \geq 2$ ML.

Some sort of InAs ''islands,'' precursors of QD's have been theoretically predicted[10] and experimentally determined[11] on the grounds of morphological measurements. These structures were referred to as platelets and quasi-three-dimensional (Q3D) structures, respectively. The present measurements clearly indicate that platelets and Q3D structures are the same nanostructures, of the same kind of those detected here and in Refs. 5 and 9. Q3D structures (or platelets), therefore, can confine carriers and give rise to bound states at energies below that of the so-called wetting layer. These structures aggregate below the critical thickness and coexist with already well-formed pyramidal QD's, at least for $L \lesssim 2$ ML.

Let us introduce now a more sensitive technique for the estimate of L_c and discuss then the critical thickness concept. PLE spectra taken in samples of the series β and δ are reported in Fig. 4. The spectra were taken at 10 K, with a detection energy equal to that of the corresponding strongest QD peak in the PL spectrum. The two main bands observed in Figs. 4(a) and 4(b) are due to recombination of photoexcited heavy-hole and light-hole excitons in the 2D InAs layer—HHE and LHE, respectively. For increasing InAs nominal coverage, the energy peak of both bands shifts to lower energies due to the increase of the average thickness of the 2D InAs layer. In the meanwhile, the linewidth increases for both the HHE and LHE bands, whose relative weights become comparable for $L \gtrsim 1.7$ ML. The latter features can be attributed to an increase in the exciton localization due to the morphological disorder induced in the 2D layer by the onset of QD aggregation. In the 1.5 ML sample of the β series in Fig. 4(a), two bands can be observed at $\sim$1.37 eV and $\sim$1.40 eV, namely, at 64 and 96 meV from the detection energy, as already reported in literature.[12] The HHE bands in the samples of the δ series in Fig. 4(b) show weak, low-energy shoulders that gain strength for $L \gtrsim 2.0$ ML. The dependence of HHE peak energy on the InAs coverage L is summarized in Fig. 5 for the two series of samples. The main peak energies are reported as solid dots and rhombuses, the

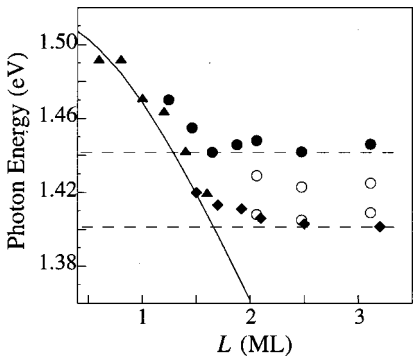

FIG. 5. Peak energy of the HHE in the wetting layer as a function of L for the samples of the β (solid rhombuses) and δ (solid dots) series. The energies of the additional shoulders observed in the PLE of δ samples are reported as open circles. Solid triangles are taken from Ref. 13. The solid line gives the HHE energy vs L as calculated for strained InAs in GaAs in a simple square-well model. The two horizontal dashed lines, which coincide with the asymptotic values of the HHE peak energy at high-L values, define the critical thickness values by their crossing with the solid line.

weak shoulder energies as open circles. In the same figure, the HHE energy evaluated in a simple effective-mass approximation for InAs ultrathin quantum well[13] is also reported as a solid line, while previous data from Ref. 5 are reported for reference as solid triangles. The HHE energy falls off sharply and coincides within the experimental uncertainty on L (see Table I) with the data from Ref. 5, as well as with the theoretically expected trend, at least for low-L values. For high-L values, the HHE energy begins to saturate and asymptotically converges to a constant value. This behavior defines an empirical value of the critical thickness for QD self-aggregation. In fact, the crossing between the theoretical curve (solid line) and the asymptotic constant value (dashed lines) defines the value L_c of L over which QD nucleation starts and the thickness of the 2D layer suddenly stops to increase. With this definition, L_c is equal to ~ 1.4 ML for the samples of the δ series, grown at 520 °C, and is equal to ~ 1.7 ML for the samples of the β series, grown at 500 °C. If thermal equilibrium conditions are not established during the sample growth, as should be expected in the case of molecular-beam epitaxy, the transition from the 2D to the 3D growth is smooth, as observed in the case of the samples of the series β, grown at 500 °C. Thermal fluctuations can also broaden the transition and, therefore, the growth of the 2D InAs layer only asymptotically reaches an equilibrium value. In the case of samples grown at 520 °C (series δ), the transition from 2D to 3D seems, instead, to be sharper. These results well agree with the conclusions recently reached on the grounds of reflectance anisotropy spectroscopy (RAS) measurements,[14] a technique very sensitive to sample growth and surface conditions. In fact, RAS measurements in InAs/GaAs heterostructures indicate that a small fraction of the incident molecular-beam fluxes are still incorporated into the 2D InAs layer for $L > L_c$. This effect is more pronounced, the lower the growth temperature is, in agreement with the results presented here. It may be worth noting that the counterpart of this effect is observed in Fig. 1, where the QD emission energy exhibits a sharper transition from region II to region III the higher the sample growth temperature is. It should be emphasized also that optical—in particular PL-based techniques—are by far more sensitive than the RHEED technique generally used to determine L_c. In fact, the former technique detects the weak emission of the carriers localized in the first formed low-density QD's, while the latter technique determines L_c on the grounds of a much higher surface density of QD's, namely, that needed to produce a sizable change in the electron diffraction pattern. Therefore, 3D structures can be detected by PL even when their density is expected to be quite small. This is shown in Fig. 2 where a slight contribution to PL from QD precursors has been measured in the case of a 1.2-ML sample.

In conclusion, the onset of QD formation definitely inhibits the growth of the 2D layer, for different values of L_c for the two sets of samples grown at different temperatures. This provides experimental evidence that the critical thickness depends on the growth temperature, in agreement with the predictions of a recent theoretical model.[6] That same model, which takes into account mass transfer and In and Ga interdiffusion during the growth process, has also predicted an exponential increase of the QD density and a dependence of L_c on the sensitivity of the experimental technique used for its determination, as suggested here. The fractional values estimated for L_c show that the 2D layer evolution is subject to strong fluctuations. These give rise to a disordered final morphology with inhomogeneous WL thicknesses, a likely source for the weak shoulders of the HHE band in the PLE spectra of the δ series.[15] This evidence together with the broadening of the PLE peaks for high-L values shows that the Stranski-Krastanow picture applies to a limiting ideal case, while a major role during the growth process is played by kinetic effects, usually not taken into account by theoretical models based on thermodynamic equilibrium conditions (see, e.g., Refs. 10 and 16, and references therein). Somewhat similar conclusions have been recently achieved on the grounds of scanning tunneling microscopy and PL measurements in the same type of heterostructures studied here, which have provided evidence that 2D clusters, Q3D clusters, and 3D islands coexist in the intermediate region II and have shown a peculiar reentrant nature in the formation of Q3D clusters.[17,18]

A wider set of nonresonantly excited PL spectra taken in samples of the γ series is shown in Fig. 6. The QD bands with emission energies lower than 1.4 eV exhibit doublet or triplet structures, as summarized in Fig. 7 where the data for the full set of all samples of the γ series are reported. An analogous behavior is generally observed in the other two series of samples, β and δ, with emission bands at energies between 1.2 and 1.4 eV. Below 1.2 eV the QD bands resume, instead, a simple, symmetrical Gaussian line shape. Similar results have often been reported in literature, where multiplets are usually ascribed either to the emission from excited states or to the simultaneous presence of many QD families of different geometry and mean size.[19–23]

In the present case, none of the multiplet components can be ascribed to emission from excited states. In fact, the intensity of all the multiplet components increases linearly for an increase in the excitation power density (P) over three orders of magnitude, starting from $P = 0.001$ W cm^{-2}. Moreover, the highest-energy component of the multiplets is often

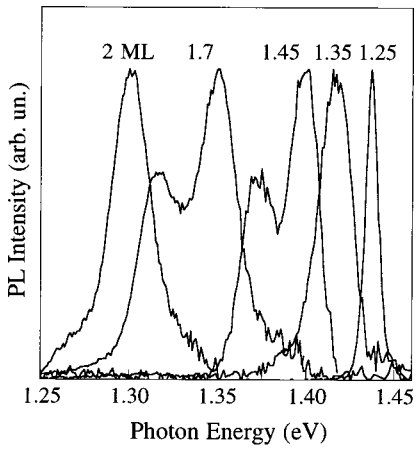

FIG. 6. PL spectra of different samples from the γ series, with L between 1.25 and 2.0 ML.

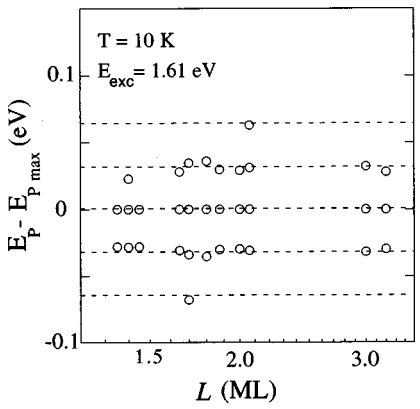

FIG. 8. The energy of each component of the multiplets observed in the PL spectra of samples reported in Table I is measured with respect to the main PL peak energy and reported vs L. The dashed lines define constant values corresponding to integer multiples of 32 meV from the main peak position.

stronger than those at lower energy, independently of the excitation power density.

The energy difference ΔE between each two adjacent components of the multiplets is roughly equal to 32 meV, within the experimental uncertainty, independently on InAs coverage or growth conditions, namely, on QD shape and size; see Ref. 3. This is shown in Fig. 8, where ΔE has been reported versus L for all the investigated samples of Table I where structured QD bands have been observed for nonresonant excitation, i.e., in samples of the series β, γ, and δ. It is highly unlikely, therefore, that those PL multiplets can be ascribed to multiple QD families with different shape or mean size.

On the other hand, a mean value of 32 meV for ΔE corresponds to the estimated and measured energy of the LO phonon in the bulk or at the interface of strained InAs, either QD or WL.[12,24–26] Therefore, we suggest that extended phonon (or local) modes and their interaction with carriers play a key role in the interpretation of the above multiplets. An inhomogeneous broadening due to strong fluctuations in QD sizes can sometimes hide the effects of that carrier phonon interaction. This leads to unusually broad QD PL bands like

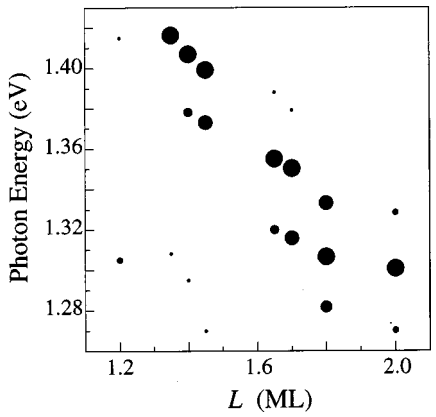

FIG. 7. Peak energies of all QD and QD precursor PL bands observed in samples of the γ series, plotted as a function of InAs coverage for nonresonant excitation conditions ($E_{\mathrm{exc}}=1.61$ eV). The relative intensity of the emission bands is proportional to the symbol size.

those observed in an energy range from 1.2 to 1.3 eV, i.e., where multiplets are more commonly observed. This last effect has been directly verified by spatially resolved PL measurements on a sample of the δ series with $L = 3$ ML. This sample exhibits fully grown pyramidal QD's and a broad Gaussian PL band under normal excitation conditions.[9] μPL measurements have been performed by using a microscope objective, which both focused the laser spot down to a diameter of 1 μm and collected the PL signal. This has allowed us to (i) increase the exciting power density by several orders of magnitude without heating the sample in order to make evident possible contributions of excited states to the PL spectra and (ii) reduce the number of excited QD's and the corresponding inhomogeneous broadening. A μPL spectrum taken at $P=10$ Wcm^{-2} in the 3-ML α sample is shown in Fig. 9 by the dotted line, together with a conventional PL spectrum taken at $P=0.05$ Wcm^{-2} in the same sample (dashed line). In both cases, a He-Ne laser has been used for the excitation, and the sample temperature was equal to 77 K. The conventional PL spectrum exhibits a single symmetrical Gaussian band, peaked at $E_0=1.267$ eV. The μPL spectrum strongly differs, instead, from the conventional PL spectrum. First, the increased power density introduces an additional band at higher energy ($E_1=1.357$ eV, 90 meV above E_0), which can soundly be attributed to a QD excited state. Second, a structured line shape can now be resolved for both the ground and the excited states, most likely because of the reduced number of excited QD's. For both states the characteristic energy spacing of the multiplets is the same, equal to 32 meV. Therefore, two multiplets, separated by 90 meV, have been fitted to the data (solid line in the figure). Each multiplet was made by three Gaussian bands 30 meV broad and spaced by 32 meV. The relative intensities of the Gaussian contributions were the only free parameters. The agreement between the model (solid line) and the data (dotted line) is quite good.

The resolution of a triplet structure in the PL of the ground state under proper condition of excitation is a feature peculiar, most likely intrinsic, of QD's emitting below 1.35 eV. In particular, this feature seems to characterize the PL spectra of the samples in the intermediate region of growth, across the critical thickness; see Fig. 6. Multiplet structures

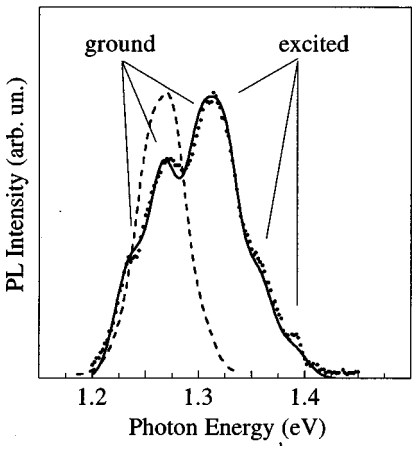

FIG. 9. Emission spectra at $T=77$ K of the $L=3$-ML α sample. Under conventional low excitation ($P=0.05$ Wcm^{-2}) PL spectroscopy a single although asymmetric band is observed (dashed line). Under 1 μm spatially resolved, high excitation ($P=10$ Wcm^{-2}) PL, that band is resolved into a triplet (dotted line). A second triplet, due to an excited-state contribution, appears in the μPL spectrum 90 meV above the ground state. The solid line gives the best fit to the data as obtained in terms of two triplets separated by 90 meV and each made of three equally spaced (32 meV) Gaussian contributions.

seem to be absent, instead, in the PL spectra of QD precursors, emitting above 1.4 eV. This has allowed us to get a rough estimate for the critical thickness also in the case of the samples of the γ series ($L_c \lesssim 1.4$ ML; see Fig. 7), whose PLE spectra do not provide clear evidence of the WL band. Multiplet structures seem to be absent also in the PL spectra of QD's emitting below 1.2 ML, namely, large QD's with high aspect ratio.[3] In the framework of an electron-phonon related mechanism, these features can be explained in terms of a dot size that maximizes the carrier charge per atom and the electron-phonon interaction strength. In fact, in QD precursors, the carrier wave function is likely spread outside the QD; in large QD's characterized by high aspect ratio values, the carrier wave function is delocalized across the whole QD, which begins to recover 3D bulk-like features. Finally, PL measurements under strictly resonant conditions in the same set of samples show that both extended modes (phonons) and modes localized in a lattice perturbed by a strong electron-phonon interaction contribute to RPL spectra.[27]

IV. CONCLUSIONS

We have shown that emission bands due to QD precursors (or Q3D's) can be observed in the PL spectra of InAs/GaAs heterostructures both before and after the onset of QD aggregation. Under suitable conditions of detection, the luminescence of Q3D's can also be discriminated from that of fully evolved QD's. Multiplet structures have been shown to characterize the PL spectra of QD in the region close to the self-aggregation critical thickness, or, more generally, of QD emitting at about 1.3 eV, as shown by microphotoluminescence measurements. These multiplets always exhibit structures separated by ~ 32 meV, the energy expected in such structures for InAs-like LO phonon modes. We ascribe the presence of these structures to effects of interaction between phonons and carriers, at least during carrier relaxation processes.

Finally, we have shown that PLE measurements on different sets of samples provide an alternative method to determine the thickness for the onset of QD formation and give an insight into the effects of kinetics during the growth. In particular, the critical thickness depends on growth temperature, and the QD nucleation freezes the WL growth. The resulting framework emphasizes the need to refine theoretical models, including kinetic effects, in order to correctly reproduce the complex growth mechanism of QD's.

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*On leave from Dept. of Physics, University of the Punjab, Lahore 54590, Pakistan.

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