Optical properties of InAs quantum dots: Common trends

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We compare results obtained in several tens of samples grown by molecular-beam epitaxy under different growth conditions with a substantial amount of data found in the literature. By plotting the photoluminescence (PL) peak energy (E_p) of the quantum dot (QD) bands as a function of the nominal thickness of deposited InAs (L) three regions are clearly evidenced in the (E_p,L) plane. Below the so-called critical thickness (L_c), three-dimensional precursors of QD's show a smooth dependence of their emission energy on L. Around L_c, QD's show a steep dependence of E_p on L, independent of the growth conditions. Finally, for $L \gtrsim 2$ ML one observes a saturation of the PL energy. This energy assumes only discrete values dependent on the growth conditions, which is attributed to the aggregation of quantum dots with different faceting.
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The interest in the properties of self-assembled quantum dots (QD's) is continuously growing because of the great potential of these structures for new devices, in particular low-threshold current lasers for optoelectronic applications. It is quite important, therefore, to understand completely the dependence of the QD optical properties and morphology on growth conditions and to gain a full control of the dot growth. These goals have not yet been achieved since the results of the large number of theoretical and experimental works on QD's seem to be model or sample dependent.

From a theoretical standpoint, models for the growth of pseudomorphic heterostructures *in thermal equilibrium condition* predict an initial two-dimensional (2D) growth. For thicknesses of deposited 2D layer L exceeding a critical value L_c, the growth evolves into the Stranski-Krastanow (SK) mode. In the case of the widely investigated InAs/GaAs heterostructure, values of L_c ranging from 1.0 to 1.8 ML have been predicted by including in the theory the mass transport, the strain relief, ripening processes, or by taking into account the evolution of stable 2D platelets into QD's.[1–5]

From the experimental point of view, a unifying picture of the results reported in the literature can be hindered by the molecular-beam epitaxy (MBE) growth conditions, which normally are far from thermodynamical equilibrium. Non-equilibrium effects may then contribute sizably to the QD formation process.[6] The QD emission energy depends on growth rates,[7] growth temperatures T_G, molecular-beam fluxes,[8] and on thicknesses, temperatures, and growth techniques of GaAs cap layers.[1,9,10] The change from 2D to three-dimensional (3D) morphology, therefore, is more subtle and complex than in a simple SK picture,[11] and experimental values of L_c reported in the literature range from-less than 1 to 1.8 ML.[12–18]

The self aggregation of 3D QD's of fixed shape but increasing dimensions should begin for $L \geqslant L_c$ and stabilize at larger L's. However, QD's may be shaped like pyramids, truncated pyramids or lenses, and their aspect ratios, namely, the pyramid height-to-base ratios, span from 0.1 to 0.6. Moreover, QD's in heterostructures with the same values of L exhibit emission bands with different peak energies E_p and line shapes, which is explained in terms of excited states, different families of dots, or multimodal distributions of dot sizes.

In this paper, we present a common key towards a better understanding of the optical properties of InAs/GaAs QD's, grown by us or in different laboratories around the world. When the ensemble of the investigated samples is considered, three regions are clearly evidenced in the (E_p,L) plane, which correspond to three different growth regimes. In the first regime, for low-L values, lens-shaped 3D precursors of QD's are deposited. In the second regime, 3D QD's begin to grow with sizes and aspect ratios rapidly increasing with increasing L. In the last regime, E_p exhibits a weak dependence on L, as in the first regime. The photoluminescence (PL) spectra, now, exhibit bands peaked at a number of discrete energies for the same amount of InAs deposited layer, *as confirmed by comparison with most of the data found in the literature*. A comparison with existing theoretical models and the analysis of the few cross-sectional high-resolution transmitted electron microscopy (HRTEM) micrographs reported in the literature suggest that those bands are due to emission from pyramidal QD's with facetings related to the growth conditions.

We have performed PL measurements on several series of InAs/GaAs structures grown by MBE or atomic layer MBE (ALMBE) (Ref. 19) in different Varian GenII chambers, under various conditions. Samples in the first series (FUB),

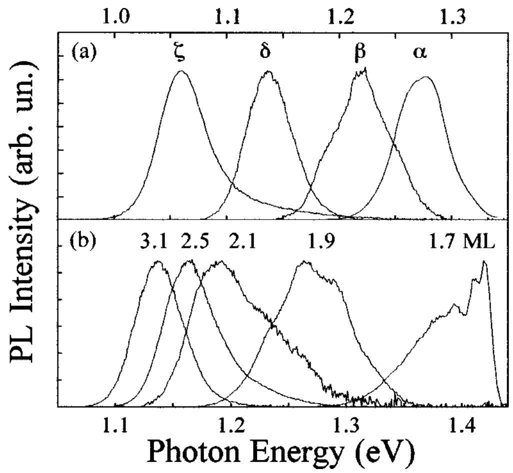

FIG. 1. PL spectra of some InAs QD's. (a) Four samples with similar InAs coverage, $L\sim3$ ML. Samples α,β, and δ have been grown by MBE under different conditions, sample ζ has been grown by ALMBE. (b) Samples from the δ series, with various values of L in regions II and III.

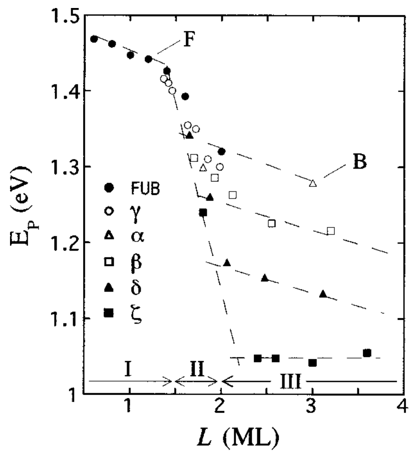

FIG. 2. Peak emission energies vs L of the main bands observed in the present work in the PL spectra of samples grown by MBE or ALMBE under various conditions. Dashed lines are guides to the eye and define three different regions in the plane. Cross-sectional HRTEM micrographs have been obtained for samples labeled by capital letters.

grown at the Fondazione Ugo Bordoni, consist of a single InAs quantum well with $0.6\lesssim L\lesssim2$ ML, grown at 420°C.[12,20] The other samples have been grown at CNR-MASPEC. Those prepared by MBE consist of QD's deposited at 500 °C (series α,β,γ) and at 520 °C (δ) and GaAs cap layers grown at 350 °C for 5 ML and then at 600 °C (α,β) or entirely at 500 °C (γ) or at 520 °C (δ). The ALMBE structures (ζ) have QD's and caps grown at 460 °C and at 360 °C, respectively.[9] The growth of part of caps at low temperatures is aimed at reducing the interaction between dots and caps. The γ samples have been obtained in a single-growth process by keeping the substrate fixed at a tilt angle with respect to the molecular beams.[13] This has allowed us to study samples with 1.2 ML $\lesssim L\lesssim$ 2 ML *but otherwise identical.* All PL measurements have been performed at 10 K, with excitation wavelengths of 488 or 780 nm.

In Fig. 1(a), we show the PL spectra of four samples, all with the same InAs coverage ($L\sim3$ ML). The peak energy of the QD band changes by about one tenth of an eV from sample to sample. The band centered at ~1.2 eV exhibits a fine structure, which seems less evident in the other bands, although they are all 50-meV wide. In Fig. 1(b), we show the PL spectra of the δ samples with L ranging from 1.7 to 3.1 ML. The PL spectra are now characterized by the presence of a few QD bands. Some band shows a fine structure, as found for the 1.2 eV band in Fig. 1(a).

The peak recombination energies of the most intense QD PL bands observed in our samples are reported in Fig. 2 as a function of L. Different symbols refer to different series of samples. Most surprisingly, *the peak energies follow a well-defined pattern,* as shown by the dashed lines, guides to the eye, which define three different regions — I, II, and III — in the (E_p,L) plane.

In region I ($L\lesssim1.5$ ML), the recombination energies depend linearly on L, with a slope of the order 40 meV/ML.

In region II ($1.5\lesssim L\lesssim2$ ML), namely, around the critical thickness, E_p changes much faster with L (about 0.6 eV/ML) than in region I and all data lie roughly on the same straight line. Under proper excitation and detection conditions, the PL spectra of the samples in region II often exhibit rich structures like those shown in Fig. 1(b) and the spectral weight shifts from higher to lower energy for increasing L.

In region III ($L\gtrsim2$ ML), the rate of change of E_p with L goes back to that observed for $L\lesssim1.5$ ML, with the exception of the ALMBE samples whose emission energy is the lowest among all investigated samples and is independent of deposited InAs layer, at least for $L\gtrsim2$ ML. Each series of samples gives rise to an independent set of emission energies. Only discrete values of the QD emission energy can be observed for a fixed L.

In Figure 3, values of E_p taken from the literature[18,21–26] have been plotted vs L. Data refer to InAs QD's grown by MBE, under the most different and uncorrelated conditions of growth. Only one sample, *A*, has been grown by low-pressure metal organic vapor phase epitaxy (LP-MOVPE).[22]

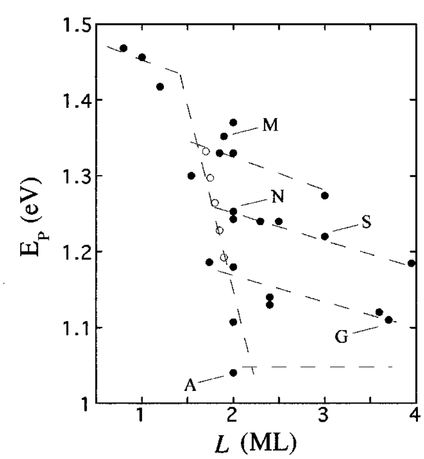

FIG. 3. Peak emission energies vs L of the main PL bands reported in the literature for samples grown by MBE, with the exception of sample *A*, grown by LP-MOVPE. Data given by open circles have been taken from Ref. 7, all others from Refs. 18,21–26. The dashed lines are exactly the same as in Fig. 2. Cross-sectional HRTEM micrographs have been reported for samples labeled by capital letters.

The dashed lines are exactly the same as in Fig. 2. The agreement between the data taken from the literature and the behavior observed for our data is excellent, thus providing a strong, additional evidence that the (E_p, L) plane is intrinsically divided into three regions, the third one being characterized by the same discrete allowed values of the QD emission energy at fixed L determined from Fig. 2. The soundness of this picture is further supported by the observation that samples with similar (E_p, L) values have also similar line shapes, i.e., the same full width at half maximum (FWHM) and band multiplicity.[27]

An intrinsic, most likely morphological, property of the InAs/GaAs heterostructure should characterize regions I, II, and III. Let us start our discussion from region III. A few of the samples reported in Figs. 2 and 3 have been characterized both optically and morphologically by cross-sectional HRTEM.[28] These samples are indicated by capital letters *B*, *F*, *A*, *M*, *S*, *N*, and *G* and their results have been reported in Refs. 20,22–26, respectively. QD's in sample *M* have (410) lateral surfaces and an aspect ratio $\sim$0.12 (0.10 in sample *B*). Samples *S* and *N* have QD's with (113) [and (114) in the case of sample *N*] lateral surfaces, while QD's in sample *G* have (110) or (320) lateral surfaces. Finally, an aspect ratio of 0.6 has been reported for sample *A*. Therefore, different dot lateral faceting characterizes the samples connected in region III by the four different dashed lines in Figs. 2 and 3. This picture is quite consistent with theoretical estimates of emission energies equal to 1.2, 1.1, and 1.0 eV for QD's with pyramidal shape, base length equal to 11.3 nm, and lateral faceting (510), (113), or (110), respectively.[29] The calculated dependence of E_p on the QD size is barely dependent on the QD shape (and likewise the dashed lines in region III are parallel), and is of the order of 32 meV/nm. This slope would correspond to a quite sharp size distribution of QD's, with a variance of 0.7 nm for a FWHM of order 50 meV, i.e., that reported for QD PL bands in region III, see Fig. 1(a).

Let us now discuss regions I and II. The PL emissions of samples with L in region I (see Figs. 2 and 3) are related to 3D precursors of QD's.[20] They are lens-shaped structures with a quite low aspect ratio, as found for sample *F*, and strongly interacting with the underlying ''wetting layer,'' (WL). The emissions related to the 3D precursors can be seen only for samples grown at the lowest temperature (420 °C) and are some tens of meV lower than those of the quantum wells associated to WL.

For L increasing in region II, the QD shape evolves from that of the precursors in region I towards those in region III defined by lateral facets whose orientations are related to the growth conditions, as discussed above. Dots in region II, therefore, have aspect ratios and PL emission energies lying between those of 3D precursors and pyramidal QD's. The steeper slope of E_p vs L in region II is related to the coincident change of both QD dimensions and shapes (instead of dimensions only, as it occurs in regions I and III). Several types of QD's can coexist in this region, as shown in Fig. 1(b). A fast change of E_p with L in region II has also been reported in Ref. 7 for samples grown by keeping the substrate fixed at a tilt angle (as done for our γ samples). For those samples, the slope of E_p vs L was decreasing with the growth rate R_G. The data reported as open circles in Fig. 3 have been obtained in samples grown at $R_G \sim 0.1$ ML/s, a value close to that used to grow our samples. In this case, the slope was roughly 0.65 eV/ML, in good agreement with present results.

Let us now make a comment on the critical thickness concept. Region II should reduce to a vertical line if the critical thickness L_c were independent of growth condition. As a matter of fact, the data reported in Fig. 2 (see β and δ series) show that L_c depends on growth temperature, at least, in agreement with a theoretical model that takes into account mass transfer.[1] Moreover, this model predicts an exponential increase of the QD density with the InAs coverage.[1] Thus, the critical thickness is an experimentally ill-defined quantity since it depends on *the sensitivity* of the technique used to measure L_c.[30]

In conclusion, a systematic analysis of the PL spectra in properly prepared sets of samples has allowed us to find well-defined trends in the QD peak emission energy vs L. These trends are common to the most part of the data reported in the literature for samples grown by MBE. Initially, for low-L values, 3D precursors of QD's start to grow. They have a lens shape, lye on a well-defined wetting layer and are capable to localize free carriers. Then, as L increases, QD's begin to develop with shapes that tend to be pyramids with facet orientations, which depend on growth conditions. Finally, for higher values of L, single families of pyramidal QD's with ''equilibrium,'' growth-dependent shapes dominate the dot ensemble. Only a discrete number of QD emission energies can, therefore, be observed for a fixed L. This picture is supported by cross-sectional HRTEM and is consistent with theoretical predictions. Finally, the lowest emission energy has been obtained, at least to the best of our knowledge, for ALMBE grown samples. Its value is independent on L and compatible with the exploitment of InAs/GaAs QD's for devices centered at the second window of optical fibers.[9]

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