Self-aggregation of quantum dots for very thin InAs layers grown on GaAs

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Low-temperature photoluminescence and transmission electron microscopy of InAs/GaAs quantum wells grown by molecular-beam epitaxy show that InAs self-aggregation of InAs quantum dots is a continuous phenomenon and that quantum dots nucleate at the well interfaces for nominal InAs layer thicknesses much smaller than commonly reported in the literature (i.e., 1.6 ± 0.1 ML). A good agreement is also found between the self-aggregated dot sizes estimated from the photoluminescence emission energies and those directly obtained from transmission electron microscopy measurements.

The presence of strain and of In segregation at the interfaces of $In_xGa_{1-x}As/GaAs$ quantum wells (QW's) grown by molecular beam epitaxy (MBE) makes the growth dynamics of these heterostructures very intriguing. Microscopic structural measurements have shown that, for increasing strain (i.e., for increasing $In_xGa_{1-x}As$ coverages) the growth of the well front changes from a two-dimensional, i.e., layer-by-layer growth, to a coherent (i.e., without generation of dislocations) three-dimensional (3D) growth, finally turning into an incoherent three-dimensional growth when strain relaxation occurs.[1–7] For molar indium concentration $x \geqslant 0.25$ and well width L greater than a critical value L_c, the coherent 3D growth gives rise to self-aggregated $In_xGa_{1-x}As$ islands,[2,3] which become confining structures for carriers after the GaAs overgrowth has been done. Since the carriers are confined along the three dimensions by the larger GaAs band gap, at all effects those islands can be regarded as self-aggregated quantum dots (SAD's). In InAs/GaAs heterostructures, atomic force microscopy[4–6] (AFM) and transmission electron microscopy[7] (TEM) have shown the high reproducibility and homogeneity of SAD's. In those heterostructures, conventional photoluminescence (PL) measurements have shown broad emission bands at energies below that of the heavy-hole free excitons (HHFE's) confined in the flat regions of InAs/GaAs quantum wells.[8,9] On the other hand, by probing sample areas smaller than $1\ \mu m^2$, cathodoluminescence and microphotoluminescence measurements[8,9] have shown that those broad bands result from the convolution of a continuum of very narrow lines with full width at half maximum (FWHM) on the order of 0.1 meV. The different emission energies correspond to the emission within SAD's of different sizes. It has been also claimed[4,5,8,9] that, if the 2D-to-3D transition is of the first order, the critical thickness L_c for the elastic relaxation of surface energy in InAs/GaAs QW's ranges from 1.5 to 1.7 ML.

Very recently, however, platelets have been foreseen to be the precursor of quantum dots[10]—and quantum wires detected[11] by scanning tunneling microscopy (STM)—for $L < L_c$. Furthermore, optical studies mainly related to HHFE recombination in InAs/GaAs quantum wells with $L < L_c$ have reported a broad PL band a few tens of meV below the HHFE band.[12–14] That band was initially associated to HHFE recombination,[12] and was later attributed to the presence of unintentional carbon impurities[13] or left unexplained.[14] Some of the present authors have already shown that a similar broad, low-energy band (band A in Fig. 1) has an intrinsic origin. The same authors have suggested that the potential fluctuations at the interfaces, associated with thickness variations around the nominal well width, localize the particles giving rise to the band A emission.[15,16]

In the present work, we have extended those previous investigations to samples with $L > L_c$, up to $L = 2.0$ ML, and successfully reanalyzed the PL spectra of the whole series of samples in terms of the model for SAD's developed in Ref. 17. Complementary TEM studies, carried out on two samples with nominal thicknesses of 1.2 and 2 ML, respectively, have confirmed the presence of SAD's in both samples. Moreover, dot sizes as estimated by applying to the PL results the same theoretical model of Ref. 17 well agree with those estimated here directly from TEM micrographs, as well as with data reported in the literature for $L \geqslant 1.6$ ML. Finally, the dot generation turns out to be a continuous phenomenon, SAD's existing already for well width thicknesses lower than the known critical value, with SAD radius smoothly increasing with the InAs coverage.

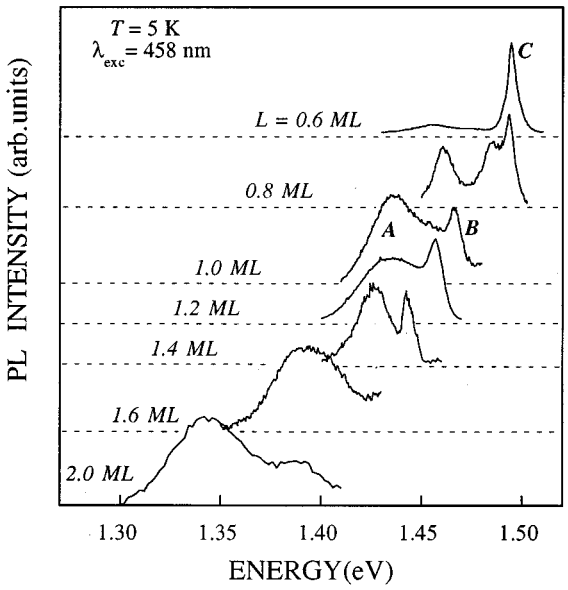

FIG. 1. Photoluminescence spectra of the seven samples used in this work. The nominal thickness of the InAs/GaAs quantum well is indicated. The band labeled *A* is due to radiative recombination in quantum dots, band *B* is the HHFE recombination in the flat region of the well, and band *C* is a carbon-related recombination in the GaAs substrate.

Seven InAs/GaAs single QW structures have been grown by MBE with a Varian GenII machine on GaAs(100) substrates. The nominal thickness of the InAs single QW's ranges from 0.6 ML to 2.0 ML. The fractional InAs coverage indicates the percentage of the sample surface covered by the first (or second) monolayer. A 0.5-μm GaAs buffer layer has been grown at 520 °C, the growth temperature being subsequently lowered to 420 °C before growing the InAs well and 10 ML of GaAs. Then, the temperature has been raised to 520 °C to grow a 40-nm-thick GaAs cap layer. The growth rate for InAs was 0.1 ML/s under As-stabilized conditions. Conventional PL spectra have been taken at different temperatures by using the 458-nm emission of the Ar^+ laser. (001) oriented plan view weak beam dark field (WBDF) TEM investigations have been performed in a JEOL 2000FX microscope on thin foils prepared by mechanochemical polishing followed by low-temperature Ar^+ ion milling in a 600 Duomill Gatan system.

The PL spectra at $T = 5$ K are reported in Fig. 1 for all the investigated samples. The incident power density is on the order of 10^2 W cm^{-2} for all samples but the 1.6- and 2.0-ML-thick QW's, which are less efficient and which have been measured at 5×10^2 W cm^{-2}. Two bands, *A* and *B*, can be observed in all samples. It has been already shown[15,16] that band *B* is mainly due to the recombination of the HHFE confined in the InAs QW. In the 0.6-ML sample, the emission of band *B* cannot be resolved from that of a carbon impurity in the GaAs substrate, namely, band *C* at 1.494 eV. The relatively small values of the PL full width at half maximum of the HHFE (about 5 meV) (Refs. 15, 16, and 18) and of the Stokes shift in the PLE measurements of deuterated samples (about 4 meV, not reported here) prove the good quality of the InAs/GaAs QW's.

Let us now discuss the band *A*. It lies a few tens of meV below the HHFE peak for all investigated samples. For increasing well widths, it shifts to lower energies and gains in relative strength until it becomes the predominant feature for $L \geqslant 1.0$ ML. No clear sign of abrupt changes in band *A* line shape or intensity can be found as a function of *L*. Different results can be found in the literature regarding this or similar bands, depending on well width values and, likely, growth conditions. A band similar to the present band *A* has been reported[8,9] for $L \geqslant 1.7$ ML. It has been attributed to SAD's at the interface between InAs and GaAs layers, in agreement with microscopic structural measurements. On the other hand, it has been already mentioned that a similar recombination band has been reported also for $L < 1.7$ ML on the low-energy side of the HHFE band.[12–16] It has been suggested[15,16] that the potential fluctuations generated by the thickness variation around the nominal well width may localize the carriers at the interfaces. Those potential fluctuations would provide the continuum of states responsible for band *A* emission. After the present more detailed and extended measurements this picture can merge with the SAD picture, *provided the self-aggregation of quantum dots begins already for L = 0.6 ML*. As a matter of fact, the band *A* observed here in the wells with thicknesses 1.6 and 2.0 ML is very similar in line shape to the spectra shown in Refs. 8 and 9 for samples of 2.0 and 2.2 ML. There, the SAD nature of band *A* has been demonstrated by means of highly spatially resolved cathodoluminescence and photoluminescence measurements, which have separated the band *A* emission into a discrete number of lines corresponding to the δ-like density of states typical of SAD's. The carrier localization at the InAs/GaAs interface, invoked to explain the band *A* photoluminescence,[15,16] can be, therefore, accounted for by SAD's with a continuous distribution of radii at the interface of the quantum well.

The redshift of band *A* for increasing nominal well thicknesses, shown in Fig. 1, suggests that the sizes of the InAs dots embedded into the GaAs increase with *L* (the carrier confinement decreases with SAD sizes). In Ref. 17, the dots are approximated by cones with a radius *r* and a base angle close to 12°, which lie on a 1-ML-thick InAs layer *d*. If we use that model, from an estimate of the energy levels in InAs/GaAs strain-induced SAD's we get the dot mean radii reported vs *L* in Fig. 2 together with the data obtained from AFM and PL measurements in Refs. 6 and 9. For thin InAs layers, the value of the SAD mean radius is roughly a constant on the order of 5 nm, then it increases for $L \geqslant 1.2$ ML. The match between our set of data and the other two sets is quite good, thus casting a first doubt on the existence of a critical thickness $L_c = 1.6 \pm 0.1$ ML in the present samples. This is further supported by TEM investigations, the results of which will be briefly discussed in the following. As an example, the TEM micrograph of Fig. 3 concerns a sample with nominal InAs coverage of 1.2 ML, i.e., less than the critical thickness. ''Cometlike'' islands of different size and length, forming an angle of about 30°–35° with one of the $\langle 001 \rangle$ directions, appear on the nominally (001) oriented GaAs substrate. Quantum-wire-like structures have been recently reported[11] on 1–2-ML-thick InAs/GaAs layers grown with short growth interruption times on exactly oriented substrates. It has been shown that those structures turned into

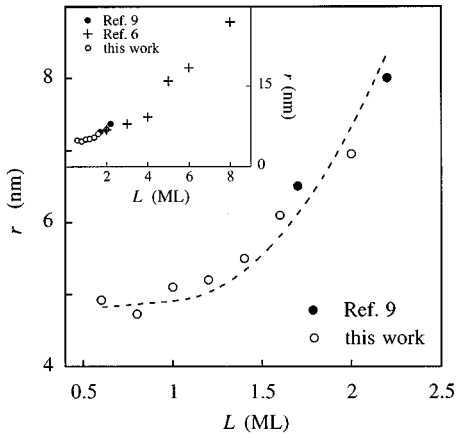

FIG. 2. Mean dot size of the SAD's as a function of the nominal thickness of the InAs/GaAs quantum well (open circle) as obtained from the analysis of the PL data. The full dots are results obtained by PL in Ref. 9. In the inset, the comparison is extended to results obtained in Ref. 6 by AFM in larger wells.

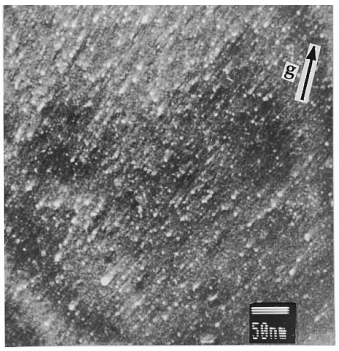

FIG. 3. WBDF (001) oriented plan view TEM micrograph of cometlike structures in a 1.2 ML nominally thick sample. Individual SAD's of different sizes are clearly shown to constitute the "comet." g-$3g$ = (002) type.

individual SAD's, only in the 2-ML sample, after increasing the growth interruption times. In this work, on the other hand, a careful analysis of Fig. 3 shows that, despite the fact that no growth interruption has been used and the well thickness is smaller than L_c, the InAs cometlike structures are actually formed by a few adjacent individual SAD's of decreasing radii.

Since the TEM analyses have been carried out in the WBDF mode, the SAD absolute sizes are close to the real ones.[19] As a consequence, it has been possible to estimate the spread of the SAD radii that ranged from 1 to 5 nm with an average value of 3 nm, in good agreement with the results of present PL studies on the same sample (see Fig. 2). Further, the selected area diffraction patterns of the InAs layers revealed that, on the average, SAD's were strained to the GaAs substrates, as also assumed in the analysis of our PL data. The same structures with SAD's of larger average radii have been also found by TEM investigations performed on a 2-ML-thick sample. A detailed description of this sample will be reported elsewhere. A rough estimate of the SAD areal density from micrographs such as that reported in Fig. 3 gives a value of about $1.5\text{-}2\times10^{10}$ cm^{-2}, in good agreement with structural measurements in thicker wells,[4,7,8,20] which estimate dot areal density ranging between 3 and 9 $\times10^{10}$ cm^{-2}. This quantity can also be roughly estimated from PL data by (i) imposing the volume conservation of deposited InAs, and (ii) assuming an equal relative probability for the formation of InAs or GaAs flat regions at the InAs/GaAs interfaces. Dot radii have been taken from Fig. 2; the flat regions have been assumed to be squares whose sides (about 3 nm) have been deduced by the analysis of the FWHM of the photoluminescence of the HHFE.[18] In this case, the dot density comes out to be $\sim 3\times10^{11}$ cm^{-2} for $L = 1.2$ ML. The different sensitivity of the PL and TEM techniques might explain those different estimates of the SAD areal density.

In summary, TEM observations well support our PL predictions. Neither TEM nor PL give evidence of an abrupt transition in the aggregation of SAD's. They provide, on the contrary, evidence of a smooth transition between two different regimes of SAD aggregation, for $L\leqslant1.2$ ML and for $L>1.2$ ML. In fact, the dot radius, initially constant with L, increases for $L\geqslant1.2$ ML, while the global luminescence efficiency increases with L up to 1.2 ML and decreases for greater values of L (not reported here).

On the grounds of the present and previous results, we suggest that a continuous picture describes the transition from the layer-by-layer, two-dimensional growth to the coherent, three-dimensional growth. For very thin InAs coverages, 1-ML-thick islands of InAs begin to nucleate with an average radius $\sim$3 nm, as indicated by the study of the FWHM of the heavy-hole free-exciton line.[18] A few of those islands, with radius on the order of 5 nm or less, and likely higher thickness, are candidates as the precursor structures recently foreseen for thin wells.[10] They may localize carriers strongly enough to give rise to a separate luminescence band, the band A. The radius of those small SAD's increases with the nominal InAs coverage, faster for $L>1.2$ ML (see Fig. 2). At the same time, the emission energy of band A begins to decrease and its separation from the HHFE band increases (see Fig. 1). For $L>1.2$ ML, the larger SAD's are more easily detected by AFM and TEM measurements. A new, even faster increase in the SAD radius can be expected for $L\sim4$ ML (see Fig. 2 and Ref. 6), i.e., when the incoherent strain relaxation starts.

In conclusion, by means of a detailed analysis of the photoluminescence of InAs/GaAs quantum wells combined with TEM measurements, we have shown that self-aggregated quantum dots nucleate for InAs layers thinner than previously reported on the grounds of structural measurements. The transition from a 2D growth mode to a 3D coherent one is continuous with nominal InAs thickness, although it develops faster for $L>1.2$ ML. The SAD average radii change continuously with the InAs coverage, starting from 5 nm, as evaluated from the energy position of a band observed at energy smaller than that of the heavy-hole free exciton. The values of the dot radii estimated from photoluminescence spectra match well those extrapolated from TEM measurements in the same samples.

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