Self-organization processes of InGaAs/GaAs quantum dots grown by metalorganic chemical vapor deposition

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$In_{0.5}Ga_{0.5}As/GaAs$ (001) quantum dots (QDs) were grown by metalorganic chemical vapor deposition (MOCVD) on exactly (001) oriented substrates using the Stranski–Krastanow growth mode. The dot density and their relative geometrical arrangement are found to be strongly dependent on the substrate temperature. The dots have identical square shaped bases oriented along ⟨100⟩. For high densities a preferential relative alignment of the dots along the ⟨110⟩ directions is found. These dots tend to be arranged in a chainlike pattern with decreasing dot size towards the ends of the chains. From these observations the dot formation process for $In_{0.5}Ga_{0.5}As$ quantum dots is suggested to be driven by energetics whereas the relative orientation is governed by kinetic effects. © *1996 American Institute of Physics.*[S0003-6951(96)00923-0]

Heteroepitaxial growth of highly strained structures like $In_xGa_{1-x}As/GaAs$ has gained increasing interest as it offers the possibility to fabricate self-organized structures like quantum dots (QDs) and quantum wires without any substrate patterning process. These low dimensional structures show unique physical properties, particularly interesting for novel optoelectronic devices like QD lasers with low threshold current density and high T_0.[1] A fundamental question for future applications is whether dot formation is kinetically driven or due to minimization of total energy. The latter leads to sharper dot size and shape distributions. Thermodynamical equilibrium calculations predict a hierarchical series of self-organization processes.[2] According to these calculations the equilibrium state for a dilute quantum dot array is an ensemble of identically shaped pyramids of square base parallel ⟨100⟩. Their equilibrium size is defined by the ratio of facet energy and strain relation at facet edges. For a dense array the interaction between the individual dots via strain fields leads to an alignment of the dots on a two-dimensional square array oriented along the ⟨100⟩ directions thus representing an even higher level of self-organization.

For QD structures grown by molecular beam epitaxy (MBE) there exists a wealth of information[3–7] including the first observation of self-organization of InAs quantum dots.[7] For MBE grown $In_xGa_{1-x}As$ dots ($0.3<x<0.6$) on singular (001) surfaces the formation of wirelike structure oriented along ⟨110⟩ has been observed.[8,9] In contrast there exist only very few reports on metalorganic chemical vapor deposition (MOCVD) grown InGaAs/GaAs QDs.[10,11] Preferential dot formation at surface steps has been observed for grown on misoriented substrates.[10] Self-ordering of dots grown by MOCVD on exactly oriented substrates has not yet been reported. In this letter, we present the first observation of self-organization of InGaAs/GaAs quantum dot structures grown by MOCVD on exactly oriented substrates. We find the dots to be aligned along the ⟨110⟩ directions in contrast to the alignment found for MBE grown InAs dots, though their base shape is identical to MBE grown dots.

The quantum dot structures of nominal In content of 50% were grown by low pressure MOCVD at 20 mbar total pressure on exactly oriented undoped GaAs (001) substrates using TMGa, TMIn, and pure AsH_3 as precursors. The growth temperature for the dots was varied between 425 and 525 °C. Following a growth interruption of 180 s a cap layer was grown at the same temperature. The nominal InGaAs layer thickness was 5 ML with a growth rate of ~0.03 ML/s. The AsH_3 partial pressure was set to ~0.2 mbar, since a comparison of dot samples grown at different AsH_3 fluxes yielded highest dot densities for moderate AsH_3 pressures. The optical and structural properties of the samples were examined by photoluminescence (PL) with excitation densities varying between 5 mW/cm^2 and 500 W/cm^2 in the temperature range from 8 to 300 K. Bright field transmission electron micrographs (TEM) were taken in a JEOL JEM1000 microscope (1 MeV acceleration voltage) using conventional diffraction contrast techniques.

Figures 1(a)–1(f) show plan view TEM images of three samples grown at 425, 475, and 525 °C. Two classes of three-dimensional objects can be distinguished. Besides the quantum dots with lateral dimensions of ~15–25 nm with similar shape to those found in MBE samples,[7,8] we find large clusters with a base length of 50–500 nm with nonuniform shape. In these clusters the strain is plastically relaxed. The dot density systematically decreases with increasing temperature. The cluster density also decreases but the cluster size is rapidly growing. From a detailed inspection of the TEM images, different origins of the clusters for the different samples can be deduced. For growth at 425 °C the statistically oriented clusters are obviously composed of coalesced dots [Figs. 1(a) and 1(b)]. The clusters in the low density 525 °C samples show a rectangular base oriented along ⟨110⟩ [Fig. 1(f)]. For this sample we also find oval shaped objects

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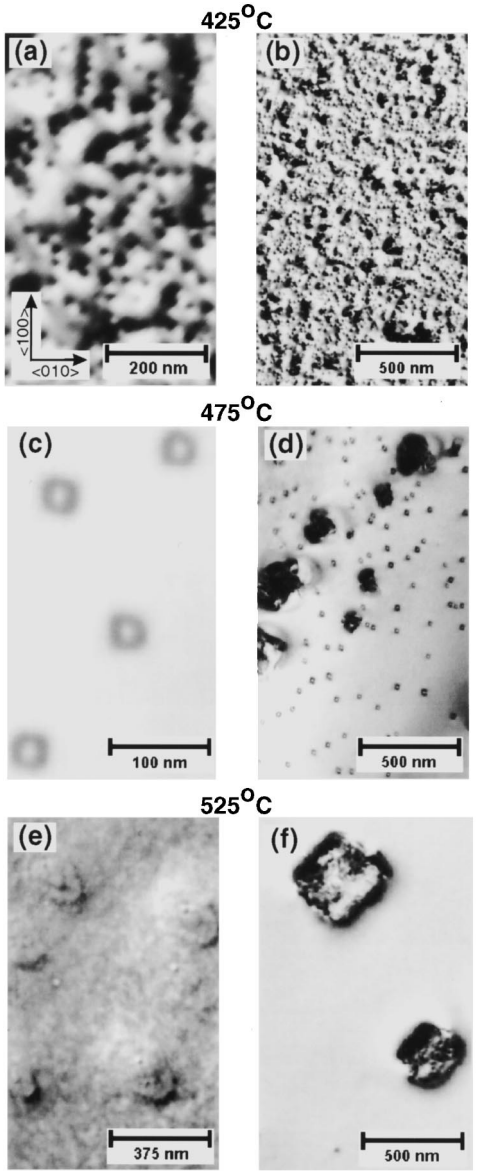

FIG. 1. Plan view TEM images of three InGaAs/GaAs dot samples grown at temperatures of 425 °C (a),(b); 475 °C (c),(d); and 525 °C (e),(f). The orientations given in (a) are the same in all pictures.

of typical dimensions of 200 nm×300 nm, which show only very weak contrast indicating a lower thickness as compared to the quantum dots in Figs. 1(a)–1(d). Most of these disklike structures contain one single dot near the center as can be seen in Fig. 1(e). The base of the coherently grown quantum dots, which are best resolved in the bright field [001] image of 475 °C sample [Fig. 1(c)], is found to be square-shaped parallel ⟨100⟩ coinciding with the findings in Ref. 8 for MBE grown dots, but in contrast to recent observations of a hexagonal dot shape for MOCVD grown InP/InGaP quantum dots.[12]

The highest dot/cluster density ratio, desirable for optoelectronic applications, was observed for low growth temperatures at moderate group V pressures of about 0.2 mbar. For such samples the relative dot positions were statistically evaluated for ~1300 dots (~3.2 μm^2). The distribution function in Fig. 2 shows a preferential alignment of the nearest neighbors in the ⟨110⟩ directions with a lower but still observable peak along ⟨100⟩. The resulting average nearest-neighbor distance is ~23 nm. It should be pointed out that the dot local density varies on a scale of about 200 nm leading to a domainlike density distribution. Thus, the large scale average density (~4×10^8 cm^{-2}) is not representative for the maximum local density, which is up to ~8.2×10^{10} cm^{-2}.

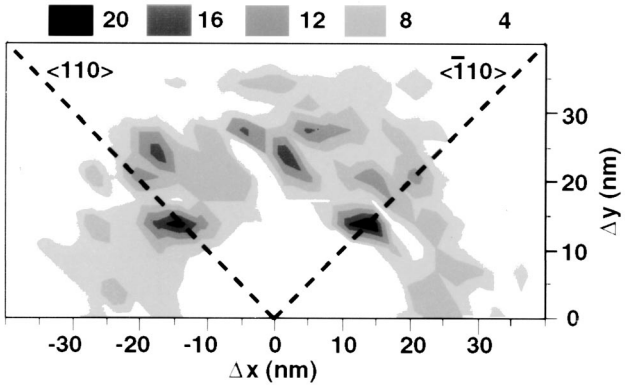

FIG. 2. Two-dimensional histogram of relative next neighbor dot positions of ~1300 dots of high density dot sample (T_S=425 °C).

These structural properties strongly affect the optical properties of the samples as shown in Fig. 3. The PL spectra of the 425 and 475 °C samples are dominated by the QD luminescence around 1.0 eV not present in the 525 °C sample. The dot luminescence in Fig. 3(a) consists of a single peak at 1.03 eV, whereas the 475 °C sample shows two lines at ~50 meV lower energies separated by 35 meV. For this sample, additional features close to the expected wetting layer luminescence energy of 1.35 eV are found. For the sample grown at 525 °C the wetting layer luminescence around 1.38 eV is dominant [Fig. 3(c)]. On the low energy side of this peak a broad shoulder is found which is typically observed for samples with a low density of fully developed dots.

A striking feature of the high density sample

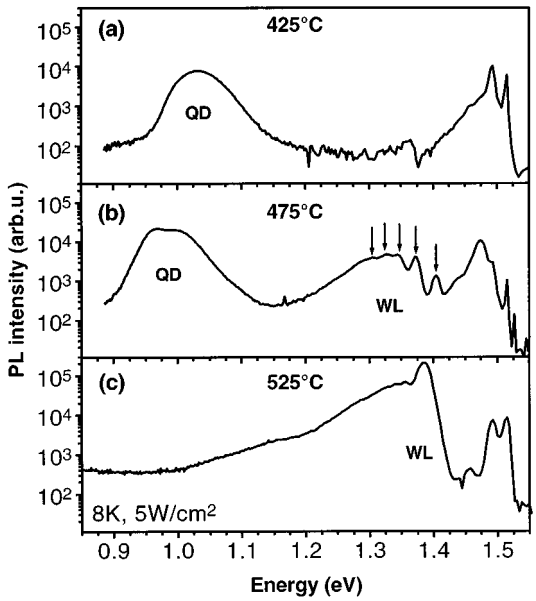

FIG. 3. Low-temperature PL spectra of the three samples shown in Fig. 1 (intensities not normalized).

(T_S=425 °C) is the chainlike dot arrangement [see Fig. 1(a)]. Most of these chains are aligned along ⟨110⟩ similar to the findings for MBE grown InGaAs QDs.[9] The dot size in such chains systematically decreases towards the ends of the chains. An influence of substrate misorientation on the dot alignment can be excluded since the misorientation was checked to be $\leqslant 0.07°$ corresponding to a mean surface step distance of about 230 nm.

This chainlike dot arrangement and the preference of the ⟨110⟩ directions are not expected from simple strain relaxation considerations that predict an alignment along the ⟨100⟩ directions parallel to the minima of the stiffness tensor. An alignment along ⟨100⟩ and the formation of a 2D square lattice is observed for MBE grown InAs dots.[7] The formation of ⟨110⟩ oriented chains indicates an increasing influence of growth kinetics with decreasing In content independent of growth mode (MOCVD and MBE). Kinetic processes govern selective epitaxy, e.g., growth on masked or patterned substrates, where the anisotropy of the diffusion length of adatoms along different crystal directions[13] and on different planes determine the shape of selectively grown structures.[14] These kinetic effects clearly determine the shape of the large clusters of the 525 °C sample. Their size is far beyond the critical size for dislocation generation. Thus the strain is relieved by plastic relaxation and a minimization of strain energy plays no role for these clusters. Instead their shape is mainly dominated by kinetics leading to the different base orientation as compared to the pseudomorphic dots shown in Fig. 1(c).

The decrease of the dot size towards the chain ends in the 525 °C sample cannot be explained by equilibrium theory either since it indicates preferential formation of new dots at ends of already existing chains. In contrast the interactions via strain fields would lead to an effective dipole–dipole elastic repulsion between neighboring dots[2] and to the formation of new chains along ⟨100⟩ directions.

The enlargement of the clusters together with the reduction of dot and cluster densities for higher T_S can be explained by an enhanced adatom surface diffusion length. This density reduction leads to a relative enhancement of wetting layer luminescence at 1.35 eV for the 475 and 525 °C samples [Fig. 3(c)]. The splitting of the 1.35 eV luminescence for the 475 °C sample [marked by arrows in Fig. 3(b)] can be attributed to luminescence from islands which differ in height by multiples of 1 ML ($a/2$ splitting). The peak energies fit to islands from 8 to 4 MLs in height with an In content of ~30% which is clearly below the intended value of 50% but agrees qualitatively with the observation of a reduced In content in the wetting layer for similarly grown InGaAs quantum dot samples.[15] The origin of the doublet around the dot energy at 0.98 eV for this sample is not yet clear. The relative intensities of the two lines do not change for excitation densities varied between 500 and 0.005 W/cm^2. Further investigation will be carried out to answer the question of whether this doublet originates from a bimodal dot size distribution or from incomplete carrier relaxation.

In summary, we have observed two different types of self-organization effects for MOCVD grown $In_{0.5}Ga_{0.5}As/GaAs$ quantum dots. The dots have identical square shaped bases originated along ⟨100⟩ and are aligned along ⟨110⟩. The latter observation is in agreement with MBE grown InGaAs dots but in contrast to InAs dots. On the other hand the base shape is identical to MBE grown InAs dots. Our results suggest that the shape of InGaAs dots is governed by a total energy minimum whereas their relative arrangement is controlled by kinetic effects.

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