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Transition from self-organized InSb quantum-dots to quantum dashes

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We have grown self-organized InSb quantum dots on semi-insulating InP (001) substrates by molecular beam epitaxy. We studied the size dependency of the uncapped InSb quantum dots on the nominal thickness of the deposited InSb by atomic force microscopy. The dot sizes have a pronounced minimum at about 2.2 monolayers of InSb. After a nominal thickness of 3.2 monolayers we observe a drastic change of the dot shape, from quantum dots to quantum dashes. From there on the dots grow in a quasicylindric shape aligned in the (110) direction. © *1996 American Institute of Physics.* [S0003-6951(96)00444-5]

Zero-dimensional systems are very interesting for novel device applications, such as quantum-dot (QD) lasers, because of the expected high quantum efficiency of the atom-like active area. Furthermore, it has already been shown, that semiconductor QDs exhibit extremely narrow photoluminescence peak widths, in the order of a tenth of a meV.[1] There have been extensive studies on the formation of self-organized quantum-dots (QD) by several growth techniques, such as molecular beam epitaxy (MBE),[1–3] metalorganic chemical vapor deposition[4] and metalorganic vapor-phase deposition.[5]

One of the remaining problems of the self-organization of the QDs is the inhomogeneity of the island sizes that broaden the observed PL peaks, and their random distribution over the surface. There have also been some successful attempts to align the QDs, e.g., by growing on one-dimensionally patterned substrates or by growth on vicinal surfaces on GaAs substrates.[5]

In this letter we present the growth of InSb QDs on semi-insulating InP (001) substrates grown by MBE in a pulsed mode, where the group V element is pulsed to enhance the surface migration.[6] The geometry of the dots was studied by atomic force microscopy (AFM). The samples were grown in a conventional solid-source MBE system equipped with valved phosphorus and antimony cells. After desorption of the InP (001) oxide at 490 °C, we grew a 500 ML thick InP buffer layer, giving a streaky (2×4) reconstruction in the reflection high energy electron diffraction (RHEED) pattern. Afterwards, the InSb layer was deposited. The surface stoichiometry during growth was controlled by measuring the surface reflection difference signal of the (110) and $(1\bar{1}0)$ directions using a HeNe laser (641.3 nm) at normal incidence to detect the absorption of In dimers at the growth front.[7] The InP buffer as well as the InSb islands were grown at a rate of 0.5 ML/s at a growth temperature of 400 °C. The growth rate was calibrated by means of RHEED oscillations.

After the deposition of 1.2 MLs of InSb we observe the typical growth mode transition from two-dimensional layer-by-layer growth to three-dimensional island growth, indicated by the onset of a spotty RHEED pattern. This transition is set on earlier than for InAs on GaAs because of the extremely high lattice mismatch between InSb and InP of 10.4%. After the formation of the InSb islands we annealed the sample for 500 s at 440 °C with a continuous Sb flux in order to obtain a more uniform size distribution. After the annealing the samples were kept under Sb flux until the temperature had fallen below 300 °C to ensure that no Sb was lost from the surface.

The AFM measurements were performed with Si_3N_4 cantilevers with a bending force constant of $k = 0.05-0.12$N/m. Electronics and software came from Nanoscope III (Digital Instruments). We took pictures from various spots on each sample, in order to detect a possible inhomogeneity of the dot sizes over the sample. Typical size differences were less than 15%.

We studied the island size and shape of the uncapped QDs by means of AFM. Figure 1 shows InSb islands of a sample with nominal thickness of 2 ML. The InSb QDs seem to be randomly distributed and have a quite homogeneous size. Their density amounts to 1×10^{10} QDs per cm^{-2}. The size distribution for the dot diameter has a mean value of 24 ± 4 nm. The height distribution has a mean value of 6 ± 3 nm (The error values given here and in the following are the standard deviations of the distributions.) The InSb QDs are smaller than, for example, InAs QDs on GaAs with a diameter of 30 nm.[8] This is basically due to the large mismatch between InSb and InP (10.4%), because the QDs, predominantly, do relax by their free surface and not by the formation of dislocations at the interface. So, it is favorable for the InSb to keep its base width small and to form high

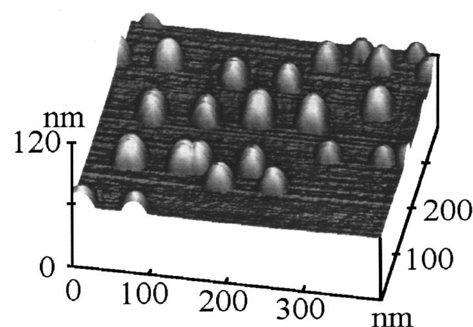

FIG. 1. AFM image of a sample with 2 MLs of InSb on InP substrate. The QDs seem to be randomly distributed and have a quite homogenous size. The density amounts to 1×10^{10} cm^{-2}. The mean diameter and height can be seen in Fig. 2.

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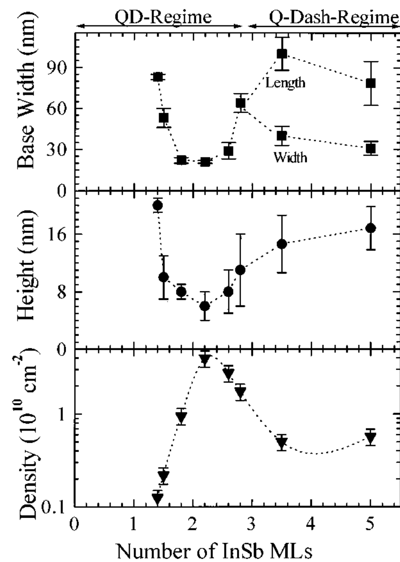

FIG. 2. Statistics on diameters, heights, and dot density for various InSb thicknesses. The statistics are made on uncapped samples by AFM. A minimum in size can be observed at around 2.2 MLs, where the density is highest. On the right-hand side the quantum-dash regime can be seen. Here the islands grow in a quasi-cylindrical shape (see also Fig. 3). The curve for the diameter divides into a curve for length and one for width, respectively. The lines are only a guide for the eye.

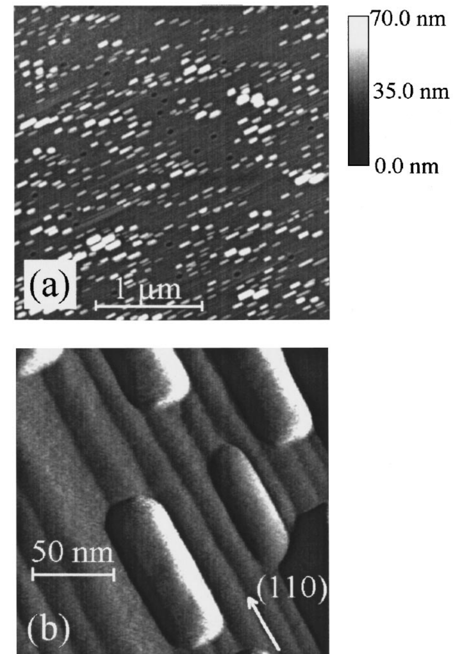

FIG. 3. AFM images of a sample with 3.5 MLs of InSb. Image (a) was taken in the height mode, while image (b) was taken in the error signal mode to emphasize the dash edges. The circular QDs have transformed to rectangular quantum dashes. The longer edge lies along the (110) direction and is about 2.5 times longer than the other one (b). On the left-hand side (a) a higher number of quantum dashes can be seen with dimensions of 100×40 nm, but there are also a few bigger ones, measuring 125×66 nm.

QDs. In fact, as compared to InAs QDs, for example, the InSb dots exhibit a smaller base width but are higher. We grew further samples from nominal InSb thicknesses of 1.4–2.8 MLs. The statistics of these samples can be seen in Fig. 2 (left part). At small nominal InSb thicknesses the dot-diameter is about 80±3 nm declining strongly with increasing number of InSb monolayers. The dot height is similarly affected by the nominal InSb layer thickness. The dot density shows a contrary behavior. As the diameter reaches its minimum at 2.2 MLs the density has a maximum of 4×10^{10} cm^{-2}. After further InSb deposition up to 2.8 MLs of InSb the dot volume rises again, while the density declines. The surprising feature is that the curve in Fig. 2 shows a minimum, that is not observed in other systems of quantum dots.

The behavior of the dot sizes and density versus the number of InSb monolayers can be explained qualitatively in the following way. After Tersoff *et al.*,[9] who calculated the formation of Ag islands on Si, an island reaches the minimal energy per unit volume at a high island density, having a quadratic shape with sidelength of a_0, given by its geometry and its chemical and elastic interaction with the substrate. So, at the onset of the 3-D growth, when the QD-density is still low, the islands grow more or less independently from each other, reaching big diameters. Because the mean inter-dot distance diminishes very quickly (about a factor of 3 within 0.4 MLs) after the first 3-D formation, the QDs interact with each other, exchanging material until they reach their optimal size, which is smaller because in this way the surface gets enhanced. One has to remember that the samples are grown in an enhanced migration mode. This optimal size seems to be reached at 2.2 MLs. Further InSb deposition, once having obtained the optimal size but a small inter-dash spacing, now has to result in new island growth, because then the QDs not only interact by material exchange but also through the elastic strain field around each dot (the InP substrate next to a QD is expanded), resulting in a repulsive force between them.[10] Therefore, having reached the minimum dot distance at 2.2 MLs, further deposition can only result in the growth of bigger dots, but not in more dots. Furthermore the density declines with bigger dots because the repulsive force rises with the dot size.

If we deposit more than 3.2 ML of InSb we observe a drastic change of the dot geometry (Fig. 3). The QDs do not have a round shape anymore, but an elongated one. After that point, the QDs maintain their quasi-cylindric shape in all samples with more than 3.2 ML of InSb forming quantum dashes (Q-dash). All Q dashes are aligned along the (110) direction and their length to width ratio is ~2.5. The main Q-dash size is 100 nm length and 40 nm width, but there are also some bigger ones with 125 nm length and 66 nm width [Fig. 3(a)]. For InAs QDs on GaAs two dot sizes have been observed for round QDs.[11] When the elongated dots form at the beginning of the annealing-step the laser-light reflected from the surface shows a strong anisotropy of factor of 20 in the (110) direction, which cannot be observed in the circular dot regime. There can also be noticed an enhanced surface roughness of the InP substrate and that the dots grow all along it. This roughness cannot be explained by the growth conditions because they were the same for all samples (Fig. 1 does not show this roughness), but might be evidence for an anisotropic strain field due to the Q-dashes. For that reason, we do not believe that the origin of this preferred growth in the (110) direction lies in the surface morphology. Our assumption is supported by the fact that the Q-dashes can grow closer to each other in the lateral than in the longitudinal

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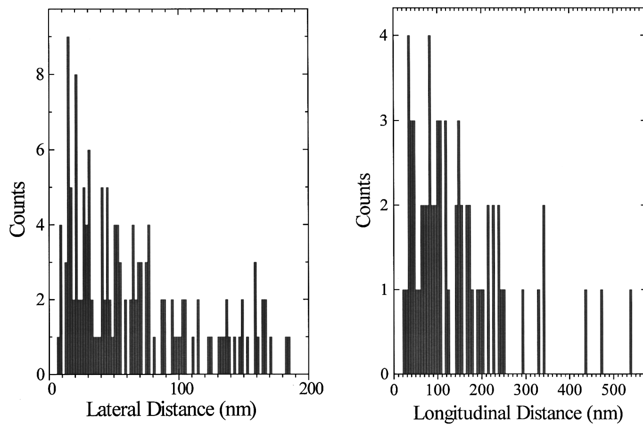

FIG. 4. Statistics on the longitudinal and the lateral spacings between quantum dashes. For the longitudinal distribution we only measured the distance of exactly collinear islands. Both the longitudinal and the lateral spacings have a minimum value of 23 and 7 nm, respectively.

direction, probably because the repulsive train field in the substrate, originated by the QDs, depends in each direction on the corresponding QD dimension in that direction.[10] The relative frequencies of the lateral inter-dash spacings **w** and the longitudinal inter-dash spacings **l** of the Q-dashes on one sample can be seen in Fig. 4. There are minimum values for w and l of 7 and 23 nm, respectively. The quotient, $l/w = 3.3$, that should depend on the square of the longitudinal and the lateral size of the islands (Ref. 10), is somewhat smaller than the theoretical value of ~ 6.3. If we suppose an exponential distribution we obtain a quotient of the mean ranges of the repulsive forces of 2, which is close to l/w, but the difference to the theoretical value could mean a slight amount of relaxation through dislocations at the interface in the (110) direction. Only a very small percentage of the Q-dashes are really oriented collinear with others, most of them are displaced in respect to their next neighbors due to the higher strain in that direction, which agrees with the picture of an anisotropic strain field. So, the minimum surface energy in the Q-dash regime is obtained with a noncollinear geometry as shown in Fig. 3(a).

According to Ref. 9, when the quantum dots pass a critical size they grow in order to minimize their energy in the dash geometry, as observed in our samples. But this model does not explain why we cannot observe ''infinite'' long islands instead of many short ones and why the (110) direction is preferred. Furthermore we can observe a distribution of the island widths, while the model predicts a constant width but the width distribution is much narrower than the length distribution, suggesting a preferred width value. The constant width in that model is also calculated at constant height, which is probably not the case as seen in Fig. 2, although the error bars are relatively large. Another explanation for the no-constant width could be that wider Q-dashes are formed by two individual ones growing together, which was actually observed in a few occasions on AFM images. In order to understand this behavior which was not explained, one has to include the different energies of the different crystal surfaces of the Q-dashes and the interaction of different islands through their strain field in the substrate material.

The initial stages of growth of InSb QDs on InP substrate have been studied by AFM. At the very first formation of the three-dimensional islands we observe very big QDs with relatively big interdot distances. At a higher numbers of InSb MLs the QDs reduce in size until they reach a minimum, from whereon they grow again. From 3.2 MLs on the quantum-dots drastically change their shape forming Q-dashes along the (110) direction. This behavior can be qualitatively understood, taking into account the elastic energy and surface energy of the QDs and the kinetics of the growth, predominantly at the island edges and the repelling force between the islands due to their anisotropic strain field in the substrate.

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[1] J.-Y. Marzin, J.-M. Gérard, A. Izrael, and D. Barrier, Phys. Rev. Lett. **73**, 716 (1994).
[2] D. Leonard, M. Krichnamurty, C. M. Reaves, S. P. Denbaar, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[3] J. M. Moison, F. Horzay, L. Leprince, E. André, and O. Vatel, Appl. Phys. Lett. **64**, 196 (1994).
[4] M. Kimatura, M. Nishioko, J. Oshinowo, and Y. Arakawa, Appl. Phys. Lett. **66**, 3663 (1995).
[5] R. Nötzel, J. Temmyo, A. Kozen, T. Tamamura, T. Fukui, and H. Hasegawa, Appl. Phys. Lett. **66**, 2525 (1995).
[6] F. Briones and A. Ruiz, Proceedings of the Sixth International Conference on MBE, La Jolla, USA, 27–31 August 1990, p. 194.
[7] Y. González, L. González, and F. Briones, J. Vac. Sci. Technol. A **13**, 73 (1995).
[8] J. M. Moison, F. Houzay, F. Barthe, and L. Leprince, Appl. Phys. Lett. **64**, 196 (1994).
[9] J. Tersoff and R. M. Tromp, Phys. Rev. Lett. **70**, 2782 (1993).
[10] L. C. A. Stoop and J. H. Van Der Merwe, J. Cryst. Growth **24**, 289 (1974).
[11] F. Bogani, L. Carraresi, R. Mattaloni, M. Colocci, A. Bosacchi, and S. Franchi, MSS7 (Seventh Intenational Conference on Modulated Semiconductor Structures), July 1995, Madrid.

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