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Self-organized lattice of ordered quantum dot molecules

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Ordered groups of InAs quantum dots (QDs), lateral QD molecules, are created by self-organized anisotropic strain engineering of a (In,Ga)As/GaAs superlattice (SL) template on GaAs (311)B in molecular-beam epitaxy. During stacking, the SL template self-organizes into a two-dimensionally ordered strain modulated network on a mesoscopic length scale. InAs QDs preferentially grow on top of the nodes of the network due to local strain recognition. The QDs form a lattice of separated groups of closely spaced ordered QDs whose number can be controlled by the GaAs separation layer thickness on top of the SL template. The QD groups exhibit excellent optical properties up to room temperature. © 2004 American Institute of Physics. [DOI: 10.1063/1.1771460]

Quantum functional semiconductor devices with applications in solid-state quantum computing and quantum communication require the lateral ordering of quantum dots (QDs) of high quality in well-defined arrangements. The self-assembly in the Stranski–Krastanov (SK) growth mode can produce QDs of high structural and optical quality which are, however, usually randomly distributed on the wafer surface. Vertical stacking of QDs can lead to lateral ordering, but for more complex QD arrangements a more precise and flexible control of the QD sites is desired. This has been mainly pursued by selective overgrowth of artificially patterned substrates which is, however, strongly limited by the spatial resolution of the lithography and/or etching steps and easily introduces additional size fluctuations and defects in the QDs. To overcome these problems, we have recently developed self-organized anisotropic strain engineering for QD ordering and demonstrated one-dimensional (In,Ga)As QD arrays of high perfection by molecular-beam epitaxy (MBE) on planar singular GaAs (100) substrates.

In this letter, we create ordered and well-separated groups of densely packed InAs QDs, lateral QD molecules, by self-organized anisotropic strain engineering of a (In,Ga)As/GaAs superlattice (SL) template on GaAs (311)B substrates. Starting from a nanoscale modulated (In,Ga)As layer, subsequent thin GaAs capping, annealing, GaAs overgrowth, and repetition in SL growth produces a highly ordered, two-dimensional (In,Ga)As-, and strain field modulation on a mesoscopic length scale. This two-dimensionally strain modulated network is utilized as a template for InAs QD growth. On top of its nodes, preferential formation of ordered groups of laterally closely spaced InAs QDs occurs to form an ordered lattice of isolated QD molecules. The number of QDs within the groups is controlled by the GaAs separation layer thickness on top of the SL template. The high structural and optical quality of the QD groups manifests itself in the excellent photoluminescence (PL) properties up to room temperature (RT).

The samples were grown by solid-source MBE on GaAs (311)B substrates. After deposition of a 250 nm thick GaAs buffer at 580 °C, the samples were cooled down to 500 °C for (In,Ga)As/GaAs SL growth. Each SL period was comprised of 3.2 nm In0.37Ga0.63As grown at 500 °C, thin capping by 0.7 nm GaAs at 500 °C, annealing for 2 min at 580 °C, and growth of a 5.5 nm GaAs spacer layer at 580 °C. The number of SL periods was between 1 and 11 in different samples. On top of the last GaAs layer of the SLs, again 3.2 nm In0.37Ga0.63As was grown at 500 °C. For the 11-period SLs, the growth was additionally terminated after thin GaAs capping and annealing, and after growth of the 5.5 nm GaAs spacer layer for structural analysis of the template evolution. The growth rates of GaAs and In0.37Ga0.63As were 0.073 and 0.116 nm/s. For InAs QD formation on the 10-period SLs, the substrate temperature was varied between 500 and 450 °C, and the GaAs separation layer thickness on top of the SLs was adjusted between 5.5 and 15 nm. InAs was deposited to a nominal thickness between 0.5 and 0.6 nm at a growth rate of 0.0013 nm/s. The structural properties of the uncapped (In,Ga)As and InAs QD layers, and SL structures were characterized by atomic force microscopy (AFM) in air and high-resolution x-ray diffraction (XRD). For the PL studies, the QDs were capped by 200 nm GaAs. The PL was excited by the 512 nm line of a Nd–YAG laser with a power density of 0.2 W/cm², dispersed by a single monochromator, and recorded by a cooled (In,Ga)As charge-coupled device.

Figures 1(a)–1(c) show the AFM images of the 3.2 nm In0.37Ga0.63As layers on the (In,Ga)As/GaAs SL templates with 1, 5, and 10 periods, respectively. For this In composition, single (In,Ga)As layers on GaAs (311)B develop a nanoscale two-dimensional surface modulation due to strain driven growth instability, rather than nucleation of QDs in the SK mode, which occurs for higher In composition. They constitute a layer of connected QDs due to the nucleationless continuous increase of the modulation height with a constant lateral periodicity. This is observed for the (In,Ga)As layer on the 1-period SL template. On the other hand, for the 5- and 10-period SL templates, distinct ordering into a two-dimensional mesoscopic mesalike arrangement is observed. For the 10-period SL template, the mesa width amounts to about 200 nm with a lateral periodicity of 300 nm and an average height of 8–10 nm.

The different steps in formation of the 11th SL period are shown in Figs. 1(d) and 1(e). After thin (0.7 nm) GaAs capping of the (In,Ga)As layer and annealing [Fig. 1(d)], a two-dimensional surface modulation with reduced height of
is reminiscent of buried translation and, hence, underlying
indicates a well-defined two-dimensional strain field modu-
layer growth periodicity of, again, 300 nm. After 5.5 nm GaAs spacer
3–4 nm is observed. The nodes are smooth with a lateral
FIG. 1. (a)–(c) AFM images of 3.2 nm (In,Ga)As on the (a) 1-, (b) 5-, and (c) 10-period SL templates. (d) and (e) AFM images of the 0.7 nm GaAs cap layer after annealing, and of the subsequent 5.5 nm GaAs spacer layer of the 11th SL period. The scan field is 2×2 μm² for all images. The black-to-white height contrast is 15 nm for (a)–(c), and 4 nm for (d) and (e).

3–4 nm is observed. The nodes are smooth with a lateral periodicity of, again, 300 nm. After 5.5 nm GaAs spacer layer growth [Figs. 1(e)], the overall surface becomes very smooth maintaining a similar modulation height. Thus, the formation mechanism of the SL template is described as follows. The nanoscale two-dimensional surface modulation of the first (In,Ga)As layer already shows a preferential ordering along the directions ±45° off [0-11], most probably due to lateral strain coupling through the substrate and/or preferential anisotropic surface migration in these directions. After thin GaAs capping and annealing, this morphology is smoothed and, again due to lateral strain coupling and anisotropic surface migration, the mesoscopic ordering builds up. It organizes itself into the well-defined mesalike structure during SL growth due to preferential (In,Ga)As growth on its nodes (indicated by the larger mesa height of 8–10 nm after (In,Ga)As deposition [Fig. 1(e)] compared to that of the GaAs spacer layer of 3–4 nm in [Fig. 1(e)], mediated by vertical strain correlation, see XRD below. The morphology is reminiscent of buried (In,Ga)As quantum disks obtained in metalorganic vapor-phase epitaxy where such a strain-driven materials reorganization is enhanced due to the higher growth temperature.13,14

When InAs is grown on the 10-period SL template of Fig. 1(e), QDs preferentially form in dense groups on top of the nodes. Figure 2(a) shows the AFM image of the QD groups formed by 0.6 nm InAs at 500 °C on the SL template. Taking the InAs QD locations as a probe of the surface strain field [the QDs preferentially form in the tensile strain field minima on top of accumulated (In,Ga)As underneath]3 indicates a well-defined two-dimensional strain field modulation and, hence, underlying (In,Ga)As distribution of the SL template. The lateral strain field modulation and strain correlated growth are directly confirmed by the XRD spec-
trum of the SL template shown in Fig. 2(b) taken with the incident beam 45° off [0-11] in the vicinity of the symmetric GaAs (311) reflection. The lateral periodicity of 350 nm derived from the satellite peak spacing of the corresponding peaks of InAs QDs grown at 470 °C on the 10-period SL template with 15 nm GaAs upper separation layer. The scan field is 1×1 μm² and the black-to-white height contrast is 10 nm for all AFM images.

The ordering and size uniformity of the QDs within the groups are significantly improved by lowering the growth temperature to 470 °C [see the 0.5 nm InAs QDs in Fig. 2(c)]. The average number of QDs within the groups is 11. A further reduction of the growth temperature degrades the ordering, while only minor influences on QD size and number are observed. A reduction of the amount of InAs mainly reduces the QD height, keeping the number almost unchanged. This indicates that the growth of the InAs QDs on the SL template follows strain-induced growth instability rather than nucleation in the SK mode, assigned to the reduced lattice mismatch on the nodes.12

The average number of QDs within the groups is controlled by the GaAs separation layer thickness on top of the SL template. The number is reduced from 11 for the 5.5 nm thick separation layer [Fig. 2(c)] to 8.5 for a thickness of 9.5 nm, and to 7.5 for a thickness of 15 nm, as shown in Fig. 2(d) for the 0.5 nm InAs QDs grown at 470 °C. The reduction of the average QD number per group with increasing upper separation layer thickness is attributed to a decrease of the tensile strain field minima, thereby decreasing the effective lateral area for preferred QD formation. Hence, by adjusting the thickness of the top GaAs separation layer, well-separated groups of ordered InAs QDs with an adjustable

number are realized on the SL template, which are themselves self-organized into an ordered lattice.

Figure 3 shows the PL spectra taken at 5 K and RT of the capped InAs QDs of Fig. 2. At 5 K, the peak at 1.18 eV with a width of 60 meV originates from the InAs QDs and that at 1.32 eV (linewidth 44 meV) from the SL template. This assignment follows from the PL peak wavelength at 1.28 eV of the SL template alone, shown in Fig. 3 for reference. The PL of the SL and QDs is well separated. The blueshift of the PL from the SL template is governed by In desorption and flattening of the (In,Ga)As layers during annealing\textsuperscript{11} [unannealed (In,Ga)As layers exhibit a peak wavelength of 1.19 eV] and varies slightly from sample to sample. At RT, the PL of the QDs is centered at 1.10 eV with a linewidth of 60 meV and a three orders of magnitude reduced peak efficiency. The absence of RT PL from the template indicates efficient carrier transfer to the QDs. Most important, the PL efficiency of the QD molecules is not degraded compared to that of InAs QDs directly grown on GaAs with a comparable linewidth.

In conclusion, ordered groups of InAs QDs, lateral QD molecules, have been formed by self-organized anisotropic strain engineering of a (In,Ga)As/GaAs SL template on GaAs (311)B in MBE. During stacking, the SL template self-organizes into a two-dimensionally ordered strain modulated network on a mesoscopic length scale due to anisotropic surface migration and strain correlated growth. InAs QDs preferentially grow on top of the nodes of the network due to local strain recognition. They form a lattice of ordered groups of QDs, closely spaced, whose number can be controlled by adjusting the GaAs separation layer thickness on top of the SL template. The QD molecules exhibit excellent optical properties up to RT making them potential candidates as functional building blocks for quantum computing and quantum communication applications in solid state.