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Complex laterally ordered InGaAs and InAs quantum dots by guided self-organized anisotropic strain engineering on shallow- and deep-patterned GaAs (311)B substrates

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Self-organized anisotropic strain engineering guided on shallow- and deep-patterned GaAs (311)B substrates is exploited for formation of complex laterally ordered architectures of connected InGaAs quantum dot (QD) arrays and isolated InAs QD groups by molecular beam epitaxy. The combination of strain and step engineerings on shallow stripe-patterned substrates transforms the periodic spotlike arrangement of the InGaAs QD arrays and InAs QD groups (on planar substrates) into a zigzag arrangement of periodic stripes which are well ordered over macroscopic areas on zigzag mesa-patterned substrates. In contrast, the formation of slow-growing facets on deep-patterned substrates produces QD-free mesa sidewalls, while InGaAs QD arrays and InAs QD groups form on the GaAs (311)B top and bottom planes with arrangements modified only close to the sidewalls depending on the sidewall orientation. The QDs on the shallow- and deep-patterned substrates exhibit excellent optical properties up to room temperature. Therefore, the concept of guided self-organization demonstrated on shallow-patterned (due to steps) and deep-patterned (due to facets) substrates is highlighted for creation of complex architectures of laterally ordered QDs for future quantum functional devices. © *2007 American Institute of Physics*.

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I. INTRODUCTION

The lateral ordering of epitaxially grown semiconductor quantum dots (QDs) with high optical and electronic quality is essential for the realization of future quantum functional devices with well-designed control of the quantum mechanical and electromagnetic interactions of single and multiple electrons and photons.[1–4] We have developed a concept for the lateral ordering of semiconductor QDs based on self-organized anisotropic strain engineering of InGaAs/GaAs superlattice (SL) templates. During SL template formation the InGaAs QD growth, thin GaAs capping, annealing, GaAs spacer layer growth, and stacking produce well-ordered one—[on GaAs (100)] and two-dimensional [on GaAs (311)B] InGaAs distributions on a mesoscopic length scale. This is caused by anisotropic adatom surface migration during annealing and vertically and laterally strain correlated growth during stacking.[5,6] The related lateral strain field modulation on the surface of the SL template governs InGaAs QD ordering due to local strain recognition.[7,8] Linear arrays of InGaAs QDs are formed on GaAs (100), whereas spotlike arrangements of connected InGaAs QD arrays and isolated InAs QD groups with controllable number of QDs are formed on GaAs (311)B, exhibiting excellent optical properties. The concept of self-organized anisotropic strain engineering for QD ordering has been extended for formation of more complex architectures of lateral QD arrays by combining it with step engineering on shallow-patterned substrates. The monolayer-height steps generated on the shallow-patterned substrates with pattern sizes of micrometer length scales modify the SL template formation to form well-positioned bends and branches of linear QD arrays on GaAs (100) and transform the spotlike InGaAs QD arrangement on GaAs (311)B into a characteristic zigzag arrangement.[9,10] The pattern sizes are clearly larger than adatom surface migration lengths ($\leqslant 1\ \mu$m) and strain field decay lengths (several 10 nm) governing the self-organization process. Hence, guided self-organization is introduced for the lateral ordering of QDs in complex architectures to create the building blocks for future quantum functional devices.

In this paper, we generalize this demonstration of guided self-organization to the lateral ordering of InGaAs, as well as InAs QDs, on shallow- and deep-patterned GaAs (311)B substrates. Similar to the InGaAs QD arrays, the isolated InAs QD groups on shallow stripe-patterned substrates are arranged in zigzags of periodic stripes which become well ordered over macroscopic areas on zigzag patterned substrates. In contrast, the formation of slow-growing facets on deep-patterned substrates produces mesa sidewalls free of QDs, while InGaAs QD arrays and InAs QD groups (coalesced into single QDs at increased growth temperature) develop on the GaAs (311)B top and bottom planes. Their spotlike arrangement is modified only close to the sidewalls depending on the sidewall orientation. The QDs on the shallow- and deep-patterned substrates exhibit excellent optical properties up to room temperature (RT) highlighting the complementary nature of guided self-organization on

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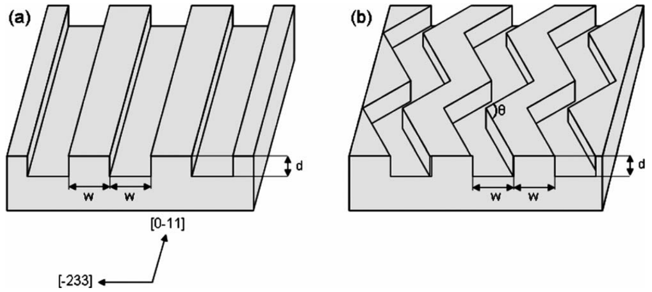

FIG. 1. Schemes of the (a) [01-1] stripe-patterned GaAs (311)B substrate with stripe widths and separations (*w*) between 2 and 10 μm and (b) zigzag-patterned GaAs (311)B substrates with 10 μm width (*w*) and angles (θ) between 60° and 150° pointing toward [−233] and [2-3-3]. The mesa depth (*d*) is 30 nm for shallow- and 200 nm for deep-patterned substrates.

shallow-patterned (large-scale modifications due to steps) and deep-patterned (local modifications due to facets) substrates for formation of complex QD arrays.

II. EXPERIMENTAL DETAILS

The samples were grown by solid-source molecular beam epitaxy (MBE) on planar and artificially patterned GaAs (311)B substrates. To study the interaction of the natural directions of ordering and adatom surface migration on planar substrates with the anisotropic growth at patterned mesa sidewalls,[11] stripe and zigzag patterns of various dimensions and orientations have been fabricated by optical lithography and wet chemical etching in the $H_2SO_4:H_2O_2:H_2O$ (1:8:1000) solution. The widths and separations of the periodic stripes, oriented along the [0-11] direction, were 2, 4, and 10 μm, as schematically depicted in Fig. 1(a), while that of the periodic zigzag patterns was 10 μm with the mesa sidewalls alternately rotated plus and minus 15°, 30°, and 60° off [0-11] [Fig. 1(b)]. The etched mesa depth was 30 nm for shallow-patterned substrates and 200 nm for deep-patterned substrates. The substrates were cleaned in concentrated H_2SO_4 and rinsed in ultrapure water before loading into the MBE growth chamber. The sample structure commenced with a 200 nm thick GaAs buffer layer grown at 580 °C, followed by a ten period InGaAs/GaAs SL template.

Each of the ten SL periods comprised 3.3 nm InGaAs grown at 500 °C, 10 s growth interruption, thin capping by 0.5(or 0.7) nm GaAs at 500 °C, annealing for 2 min at 600 °C under As_4 flux, and growth of a 5.5(or 5.3) nm GaAs separation layer at 600 °C. The In composition was maintained between 40% and 45%. On top of the SL template, the 3.3 nm InGaAs layer was repeated without annealing for formation of connected QD arrays, or 0.6 nm InAs was deposited at 480 °C (here, the thickness of the last GaAs spacer layer was increased to 15 nm) for formation of isolated QD groups. Single InAs QDs were formed by increasing the growth temperature of the SL template and InAs QDs by about 30 °C. The growth rates of GaAs and InGaAs were 0.073 and 0.132 nm/s, respectively, whereas that of InAs was 0.0013 nm/s. The As_4 beam flux was maintained between 1.8×10^{-6} and 2.4×10^{-6} Torr. The structural properties of the samples were characterized by tapping-mode atomic force microscopy (AFM) in air. A series of InAs QD samples was capped by 100 nm GaAs for photoluminescence (PL) measurements. The PL was excited by the 512 nm line of a Nd:yttrium aluminum garnet laser with excitation power density of 256 mW/cm^2. A He-flow cryostat was used to control the temperature between 5 K and RT. The PL was dispersed by a single monochromator and recorded by a cooled InGaAs linear array detector.

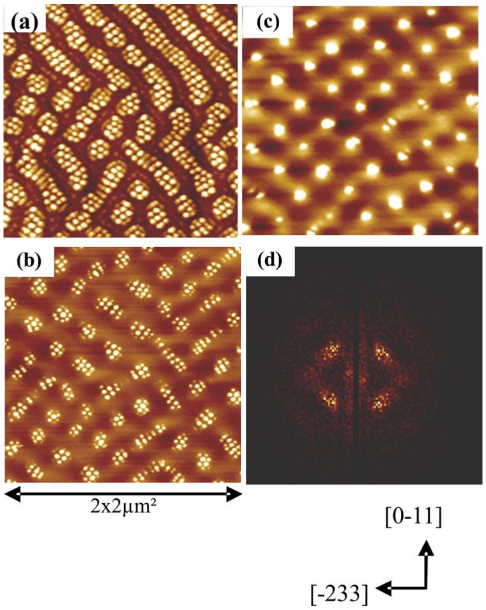

FIG. 2. (Color online) AFM height images of the (a) InGaAs QD arrays and (b) InAs QD groups on the InGaAs/GaAs SL template on planar substrates. (c) Single InAs QDs formed at increased growth temperature. (d) FFT of the AFM image of the single QDs in (c). The AFM height contrast is 20 nm.

III. RESULTS AND DISCUSSION

A. InGaAs and InAs QD ordering on planar GaAs (311)B substrates

For clarity, we briefly recall the SL template formation as well as InGaAs and InAs QD ordering on planar GaAs (311)B substrates, as described in Refs. 9 and 12. During SL template formation, the randomly distributed InGaAs QDs, after the first layer, develop into a distinct mesalike modulated InGaAs layer on a mesoscopic scale with increasing number of SL periods, generating the corresponding lateral strain field modulation on the SL template surface for QD ordering. Well-ordered periodic spotlike arrangements of connected InGaAs QD arrays and isolated InAs QD groups are formed on the SL template surface, which are aligned along the directions plus and minus 45° off [0-11], as shown in Figs. 2(a) and 2(b). The average base diameter of the InAs QDs within the groups is 35 nm and their number varies between 5 and 7. The number can be decreased to a single QD, shown in Fig. 2(c), by increasing the growth temperature. The lateral periodicity of the single QDs is 350 nm, determined from the fast Fourier transform (FFT) analysis shown in Fig. 2(d), which is similar to that of the InAs QD groups. The areal density of the single QDs is 8.5 μm^{-2}.

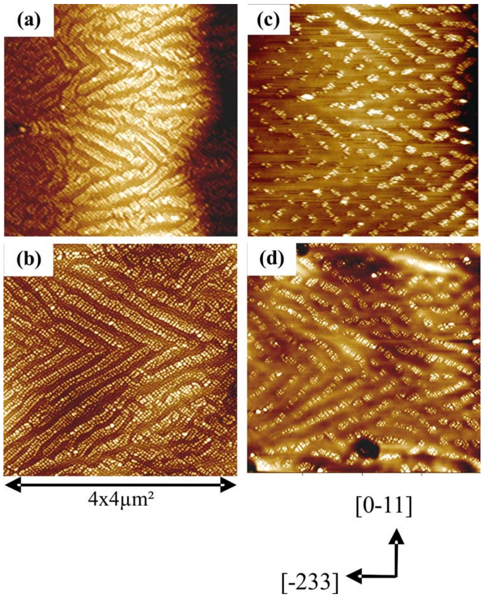

FIG. 3. (Color online) AFM height images of the [(a) and (b)] InGaAs QD arrays and [(c) and (d)] InAs QD groups on shallow-patterned substrates: [(a) and (c)] periodic stripes with 2 μm stripe width and [(b) and (d)] zigzag with acute angle (mesa bottom) of 60° pointing toward [2-3-3]. The height contrast is 30 nm.

B. InGaAs and InAs QD ordering on shallow-patterned GaAs (311)B substrates

The lateral ordering of the InGaAs QD arrays and InAs QD groups distinctly changes when the SL template is grown on the shallow-patterned GaAs (311)B substrate. Figure 3 shows the InGaAs and InAs QD layers on the stripe- and zigzag-patterned substrates with stripe width of 2 μm and acute zigzag angle of 60°. These patterns are chosen to show the most pronounced effects on the QD ordering, with the nonequivalent sidewalls of the [0-11] stripes most effectively breaking the symmetry of natural QD ordering and the 60° zigzag sidewalls supporting the natural kink structure of the GaAs (311)B surface, discussed below. On the stripe-patterned substrate the InGaAs QD arrays are arranged in periodic, well-ordered, and connected stripes assuming a zigzag pattern (on the inclined plane of the mesa stripes), shown in Fig. 3(a). The arrangement becomes uniform over macroscopic areas on the zigzag pattern (on the inclined plane with acute angle of the mesa bottom toward [2-3-3]), shown in Fig. 3(b). The InAs QD groups, depicted in Figs. 3(c) and 3(d), follow the same arrangement of zigzags of linear stripes while remaining well isolated.

The stripes of InGaAs QD arrays and InAs QD groups follow the direction of typical triangular shaped defect structure for the (311)B surface orientation with acute angle pointing toward [2-3-3], which is toward the next [111] plane.[11] Hence, the underlying kink structure of the steps induced on the inclined surface of the shallow mesa sidewalls (the inclination is less than 1°) is assigned to govern the direction of In and Ga adatom surface migration in SL template formation during annealing, and thus, the direction of the lateral strain field modulation leading to the zigzag stripelike arrangement of InGaAs QD arrays and InAs QD groups on the shallow stripe- and zigzag-patterned substrates.

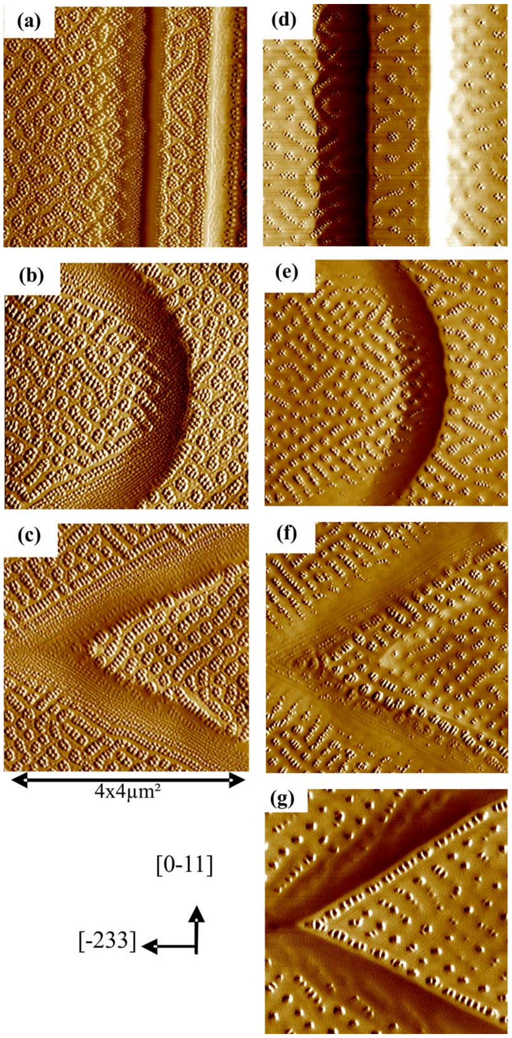

FIG. 4. (Color online) AFM slope images of the [(a)–(c)] InGaAs QD arrays and [(d)–(f)] InAs QD groups on deep-patterned substrates: [(a) and (d)] periodic stripes with 2 μm stripe width, [(b) and (e)] zigzag with acute angle (mesa bottom) of 60° pointing toward [2-3-3], and [(c) and (f)] zigzag with acute angle (mesa top) of 60° pointing toward [−233]. (g) Single InAs QDs on deep-zigzag-patterned substrate with acute angle (mesa top) of 60° pointing toward [−233]. AFM slope images are shown to reveal the QDs.

C. InGaAs and InAs QD ordering on deep-patterned GaAs (311)B substrates

In contrast to the lateral QD ordering on the inclined stepped surfaces on the shallow-patterned substrates, the large-scale ordering on the GaAs (311)B top and bottom surfaces on the deep-patterned substrates remains unchanged, as illustrated in Fig. 4. The formation of slow-growing side facets[11] produces areas which are free of QDs, and the ordering of the InGaAs QD arrays and InAs QD groups on the GaAs (311)B top and bottom surfaces is modified only close to the side facets [Figs. 4(a) and 4(d)]. This is attributed to the selectivity of growth on faceted surfaces leading to preferential adatom surface migration from the slow-growing side facets to the mesa top and bottom areas. The top and bottom areas remain planar at distances longer than the adatom surface diffusion length (of the order of 1 μm) away from the side facets, while within this distance, the growth rate enhancement produces inclined surfaces with steps to modify the QD ordering similar to the steps generated on

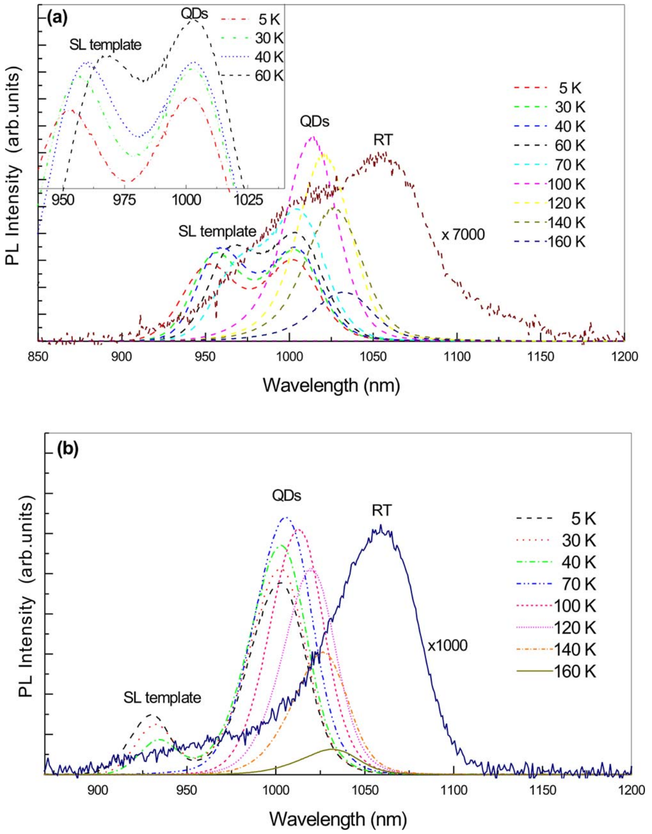

FIG. 5. (Color online) Temperature dependent PL spectra of the InAs QD groups on (a) shallow- and (b) deep-patterned substrates.

shallow-patterned substrates. Depending on the orientation of the side facets, the InGaAs and InAs QDs arrange in stripes of connected InGaAs QD arrays and isolated InAs QD groups close to the side facets, which are most pronounced on zigzag patterns with acute angle toward [2-3-3], shown in Figs. 4(b) and 4(e) or [−233], shown in Figs. 4(c) and 4(f).

The combination of self-organized anisotropic strain engineering with growth on patterned substrates is directly applied to produce complex arrangements of single InAs QDs. On the deep-patterned substrate the QDs order into single-QD stripes along the zigzag edges on the mesa top, shown in Fig. 4(g). Hence, while shallow patterns allow modifications of the QD ordering within large areas, the deep patterns allow local modifications, revealing the complementary nature of both approaches for formation of complex QD arrays.

IV. OPTICAL PROPERTIES OF INAS QD GROUPS ON THE PATTERNED SUBSTRATES

Figure 5 shows the temperature dependent PL spectra of the InAs QD groups on the (a) shallow- and the (b) deep-patterned substrates. At 5 K the PL line centered at 1000 nm stems from the InAs QD groups and that at shorter wavelength (950 nm on shallow- and 925 nm on deep-patterned substrate) from the SL template. With increasing temperature, the PL intensity from the QDs increases between 5 and 60–70 K due to thermally activated carrier transfer from the SL template to the QDs on both substrates. On the shallow-patterned substrate the PL intensity of the SL template first increases between 5 and 60 K before it drops, as depicted in the inset of Fig. 5(a). In contrast, on the deep-patterned substrate the PL intensity of the SL template decreases from the lowest temperature on, similar to the behavior on unpatterned substrates.[12] The energy separation between QDs and SL template PL is smaller on the shallow-patterned substrate, leading to thermal repopulation of the SL template at RT. These differences are attributed to the presence of steps and the resulting modification of the SL template on the shallow-patterned substrate. The steps of varying direction and density modify the QD ordering due to the local modification of the In and Ga adatom surface migration during annealing, which likely produces lateral thickness and composition fluctuations in the SL template. This leads to stronger carrier localization-delocalization effects within the SL template itself, initially increasing the template PL intensity with temperature, and to a redshift of the PL originating from regions of largest thickness and In composition. Between 70 K and RT the PL efficiency of the QDs drops about three to four orders of magnitude due to thermal escape of carriers to the GaAs barriers. This is very similar to the behavior of the InAs QD groups on planar substrates evidencing no degradation of the optical quality of the QD groups on the patterned substrates.

V. CONCLUSION

In conclusion, we have studied guided self-organized anisotropic strain engineering for formation of complex laterally ordered InGaAs QD arrays and InAs QD groups grown on shallow- and deep-patterned GaAs (311)B substrates by MBE. The combination of strain and step engineerings on shallow stripe-patterned substrates transforms the periodic spotlike arrangement of the InGaAs QD arrays and InAs QD groups (on planar substrates) into a zigzag arrangement of periodic stripes which are well ordered over macroscopic areas on zigzag mesa-patterned substrates. In contrast, on deep-patterned substrates, the formation of slow-growing facets produces mesa sidewalls which are free of QDs. The arrangements of the InGaAs QD arrays and InAs QD groups on the GaAs (311)B top and bottom planes are modified only close to the sidewalls depending on the sidewall orientation. The QDs on the shallow- and deep-patterned substrates exhibit excellent optical properties up to room temperature. Therefore, the concept of guided self-organization demonstrated on shallow-patterned (large scale modification due to steps) and deep-patterned (local modification due to facets) substrates is established for creation of complex architectures of laterally ordered QDs for future quantum functional devices.

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