Surface Structure of Manganese Gallium Quantum Height Islands on Wurtzite GaN(000$\bar{1}$) Studied by Scanning Tunneling Microscopy

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Abstract

Submonolayer deposition of manganese on gallium-rich, nitrogen polar GaN(000$\bar{1}$) surface using radio-frequency nitrogen plasma molecular beam epitaxy leads to the spontaneous formation of manganese gallium into two distinct quantum height islands, 5-layer and 6-layer islands. Atomically resolved scanning tunneling microscopy reveals the atomically flat, but unstable 5-layer island surface, and the 6-layer island surface with relatively stable row structures. We propose possible surface models for these islands' surfaces and discuss the clear structural differences explained with strains and partial relaxations. It is found that the 5-layer islands form under lateral strains and a relaxation process leading to non-uniform alternating strains results in the more energetically favorable row structures on the 6-layer island.

Keywords: manganese gallium; gallium nitride; scanning tunneling microscopy; molecular beam epitaxy

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I. INTRODUCTION

The spontaneous formation of well-defined metallic nanostructures on various Group III-V and IV semiconductors is of great interest and leads to the discovery of one-dimensional and two-dimensional nanostructures in several metal-on-semiconductor systems. One of the earliest examples is two-dimensional island structure of Ag grown on GaAs(110) surface using a two step growth procedure (low-temperature deposition followed by annealing).[1] It is driven by the quantum size effect (QSE), which plays an important role when the dimension of the metallic film is reduced to the range of the electron Fermi wavelength, and the growth is referred to as "electronic" growth.[2, 3] This specific growth mode has been observed in the Pb/Si(111) system, in which QSE-driven phenomena are observed.[4–7] Recently, manganese gallium (MnGa) is reported to form quantum height islands with two distinct heights (0.93 and 1.13 nm) on N-polar *wurtzite*(*w*)-GaN(000$\bar{1}$) surface.[8] The clear morphological difference on the surfaces between those islands, and the abrupt transformation of the 0.93 nm thick island into the 1.13 nm thick island, are interpreted. This indicates a large difference in diffusion barriers, thus implying QSE-driven growth. The islands are formed upon deposition of a submonolayer of Mn, which corresponds to the initial, transitional stage of the growth of δ-MnGa on N-polar *w*-GaN(000$\bar{1}$).[9]

The δ-MnGa is a good candidate for spintronic applications due to its high magnetic moment ($\sim$2.51 μ_B per Mn atom)[10] and Curie temperature (T_C) above room temperature.[11] An important requirement for a spintronic ferromagnet on semiconductor system is to have a sharp interface with no so-called "dead layer" or randomly intermixed non-magnetic layer. Lu et al. have reported the high quality epitaxial growth of ferromagnetic layers of δ-MnGa with ideal lattice matching and sharp interface, having a (111) orientation to *w*-GaN(0001) or (000$\bar{1}$) and that the magnetic properties are tuned by adjusting the reconstruction induced by the change in Mn flux during growth.[12] Further recent investigation on this δ-MnGa/GaN system reveals that in both simulated and experimental scanning tunneling microscopy (STM) images, 1 $\times$ 2 and 2 $\times$ 2 surface reconstructions are observed, and Ga atoms protrude more than Mn atoms and appear brighter.[13]

In this work, we further study the quantum height MnGa islands spontaneously formed on N-polar *w*-GaN(000$\bar{1}$) by molecular beam epitaxy (MBE)[8] and present recent progress on surface structures obtained using STM. Since the quantum height MnGa islands grown on

GaN are an intermediate phase in the transition to the well studied δ-MnGa/GaN bilayer system, we propose possible surface models and show that the surface structure can be understood in terms of the (111) face of the well-known $L1_0$ crystal structure for δ-MnGa.[9, 12, 13] We also discuss how that the surface structures of the 5-layer and 6-layer height islands give additional information relating to the islands' stabilities.

II. EXPERIMENTAL METHOD AND PROCEDURE

The experiments are performed in a custom-designed ultra-high vacuum (UHV) STM combined with MBE system. The MBE chamber is equipped with Ga and Mn effusion cells, a radio-frequency (rf) nitrogen plasma source, and a reflection high energy electron diffraction (RHEED) for real time monitoring of the growth surface. A quartz crystal thickness monitor is used for flux calibration of the effusion cells.

N-polar wurtzite GaN($000\bar{1}$) substrates onto which MnGa quantum islands are grown, are prepared using commercially available sapphire [Al_2O_3(0001)] substrates. After cleaning in acetone and isopropanol, the sapphire substrates are introduced into the UHV-MBE chamber and heated at $\sim$1000°C under nitrogen plasma for 45 min, then cooled down and kept at $\sim$500°C for a GaN buffer layer growth for $\sim$5 min. Following the buffer layer growth, N-polar GaN($000\bar{1}$) is grown at $\sim$650°C for 1 hr under Ga rich growth conditions to achieve a smooth surface with a thickness of $\sim$2000 Å. The vacuum pressure is kept at $\sim 2\times10^{-5}$ Torr under nitrogen plasma during the growth.

Following the N-polar GaN substrates preparation, the substrate temperature is set to $\sim$250°C for Mn deposition. During the Mn deposition, RHEED patterns are carefully monitored. The amount of Mn deposited for the MnGa quantum height islands formation is $\sim$0.2-0.6 ML, which is similar to the amount of available Ga at the surface (including both Ga adatoms and the Ga adlayer). After growth, the sample is transferred *in situ* to the adjacent analysis chamber for STM measurements. The base pressure of the STM chamber is as low as $\sim 4\times10^{-11}$ Torr. W tips are used in the STM experiments after solvent cleaning *ex situ* and electron bombardment cleaning *in situ*. All STM measurements are performed at room temperature.

III. RESULTS AND DISCUSSION

The streaky RHEED patterns in Figs. 1 (a) and (b) indicate a smooth starting GaN substrate surface, and the $\frac{1}{3}$ and $\frac{2}{3}$ order streaks indicate presence of Ga adatoms, which are typical for a N-polar GaN surface after growth under Ga rich condition, indicating a highly ordered reconstruction.[14, 15] It is understandable that these Ga adatoms are sources of Ga to form MnGa quantum height islands. Figures 1 (c) and (d) represent RHEED patterns taken right after the MnGa island formation. RHEED analysis suggests a pseudomorphic growth mode of these MnGa islands on the N-polar GaN surface, since the primary steaks remain the same before and after the island formation in both azimuthal directions, $[11\bar{2}0]$ and $[10\bar{1}0]$ of GaN. The case shown here is completely different as compared to the case of the ultrathin MnGa films grown on Ga-polar GaN, in which a 30° lattice rotation is observed within the first monolayer of growth.[9, 12, 13] As pointed by arrows in Figs. 1 (c) and (d), additional half order streaks emerge along $[11\bar{2}0]$ and $[10\bar{1}0]$. These dotted 2× patterns are indicators of the quantum height island formation and pointing out a 2×2 reconstruction on the surface. It can be observed that the MnGa island surface has 2× periodicity compared to the GaN substrate surface.

In general, MnGa crystal grown on wurtzite GaN has a δ-phase, face-centered tetragonal (fct) CuAu type-I ($L1_0$) crystal structure with a (111) growth face, and its in-plane lattice constants are $a_1 = 2.67$ Å and $a_2 = 2.76$ Å.[12] Despite the fact that these lattice constants have a huge lattice mismatch with in-plane GaN spacing of 3.189 Å, MnGa islands form pseudomorphically following the GaN lattice spacing. Depicted in Fig. 1 (e) is a model of the interface of the MnGa island and the GaN substrate, where Mn atoms replace Ga atoms within the Ga adlayer upon Mn deposition and the island formation starts under huge strains. An epitaxial relationship of the MnGa(111) islands to the GaN$(000\bar{1})$ substrate is derived with $[1\bar{1}0]_{MnGa} \parallel [11\bar{2}0]_{GaN}$.

Presented in Fig. 2 is a 200 nm × 120 nm 3D rendered STM image that reveals many interesting features of the MnGa quantum height island structures spontaneously formed on the N-polar GaN$(000\bar{1})$ surface. It can be clearly seen that the islands (green) are hexagonal in shape with flat tops and steep sides all around, and standing out of the GaN wetting layer surface (blue). The edges of the islands orient along GaN $[11\bar{2}0]$ directions, which indicates a strong anisotropic diffusion during the formation. According to our previous study, the

islands have two distinctive heights, 5-layer islands of ∼0.93 nm high and 6-layer islands of ∼1.13 nm high.[8]

Figure 3 (a) is a 40 nm × 40 nm zoom-in view of the central region of Fig. 2, containing both a 5-layer island and a 6-layer island. The islands are confirmed to be a 6-layer island and a 5-layer island based on their heights and the presence or absence of atomic chains.[8] The surface of the 6-layer island reveals a well-ordered row structure with bright and dark alternating rows running along MnGa[$1\bar{1}0$], corresponding to the GaN[$11\bar{2}0$] direction, as well as long atomic chain structures sitting on the dark valleys on its surface. No chains and less orderly row structures are seen on the surface of the 5-layer island. It is obvious that each island has a different surface reconstruction. On the corner of the 6-layer island as marked with a circle in Fig. 3 (a), a part of the 5th layer is exposed to the surface. Its height difference from the top surface is measured to be 1.9 Å, corresponding to the height difference between 5-layer and 6-layer islands in ref. [8]. It can be implied that the total area of the 6th layer is reduced, compared to the underlying layers, which is related to the strain relaxation in the 6th layer as shall be shown.

As can be seen from the 3D rendered zoom-in STM images in Figs. 3 (b) and (c), distinct surface structures are observed for each island surface. Fig. 3 (b) is the zoom-in view on the 6-layer island surface revealing more uniform and zig-zag row structures with steeper and deeper valleys compared to those on the 5-layer island surface as seen in Fig. 3 (c). The color-scale is 0.9 Å for the 6-layer island, 3× larger than for the 5-layer island. Moreover, the valleys on the 5-layer island surface are not consistent due to protrusions randomly occurring, and the row heights are inconsistently varying as seen in Fig. 3 (c).

Starting from the GaN($000\bar{1}$) surface lattice, the surface models of the 5-layer and the 6-layer islands are derived and illustrated in Fig. 4. For clarification, the Ga atoms in GaN substrate and MnGa islands are illustrated in gray and black, respectively. Fig. 4 (a) shows the Ga adlayer lattice of N-polar GaN, where the MnGa island formation takes place, and the primitive δ-phase MnGa (111) lattices. In-plane lattice constants are a = 3.189 Å for GaN, a_1 = 2.67 Å, and a_2 = 2.76 Å for δ-phase MnGa. There is a huge lattice mismatch (−0.52 Å) between MnGa(111) and GaN($000\bar{1}$). Manganese deposition on the GaN substrate at ∼250°C results in the replacement of Ga atoms within the Ga adlayer by Mn atoms, as shown in recently published paper.[16] During this process the lattice constant of GaN maintains the same according to the RHEED pattern. MnGa islands start to form

in pseudomorphic growth mode.

Depicted in Fig. 4 (b) is the top view model of the MnGa(111), showing alternating rows of Mn and Ga atoms with a 1 × 2 unit cell, corresponding to the additional 2× streaks emerging along [11$\bar{2}$0] as Mn deposition continues, as marked with arrows in Fig. 1 (c). From the huge lattice mismatch between MnGa(111) and GaN(000$\bar{1}$), this 1 × 2 unit cell model of the MnGa island can be assumed to be under lots of strains and in a metastable state. It is anisotropically strained by 15.6 % (along [1$\bar{1}$0]) and 20.5 % (along [11$\bar{2}$]), and it agrees with the lattice spacing of GaN exactly. This lateral expansion is accompanied by an out-of plane lattice contraction; as reported in ref. [8] an out-of plane lattice spacing of the islands was measured to be 1.9 Å which compared to the layer spacing of MnGa along [111] (2.2 Å) corresponds to an out-of plane strain of −13.6 %. Therefore, we find in-plane expansion to match the GaN lattice is compensated by the out-of plane contraction. It is very different from the ultrathin MnGa films grown on Ga-polar GaN which are unstrained in any direction due to a 30° lattice rotation compared to the lattice of GaN.

In general, when a Mn:Ga ratio is near or larger than 1.2:1, 2 × 2 surface reconstruction occurs by replacing every other Ga atom with a Mn atom.[12] Apparently, the flux ratio in this study falls into that range and 2 × 2 surface reconstruction can be expected to appear on the surface of the islands. This reconstructed surface model with 2 × 2 unit cell is presented in Fig. 4 (e) showing Mn rows and Mn-Ga rows. In fact, zooming-in on the 5-layer island surface as presented in Fig. 4 (d), reveals this 2× periodicity with a 6.3 ± 0.3 Å spacing between two bright atomic features along a Mn-Ga row. As presented in Fig. 4 (d) by overlaying this 2 × 2 reconstructed surface model, it is found that the Ga atoms (black spheres) are superimposed exactly on the bright atomic features along Mn-Ga rows. Appearing brighter for Ga atoms in the STM image agrees well with the result in ref. [13]. As reported in ref. [8], there are alternating variations of the row brightness (height) as indicated by white and black arrows on the top of Fig. 4 (d). Presumably, this alternating variation in the brightness (height) of Mn-Ga rows results from the huge strain.

Atomic row structures are clearly seen along GaN [11$\bar{2}$0] azimuth (corresponding to the MnGa [1$\bar{1}$0]) and the atomic corrugation is also measured along the row. The line section A given in Fig. 4 (c) is a measurement along a bright Mn-Ga row, as drawn with a red line in Fig. 4 (d), corresponding to the 3D image as shown in Fig. 3 (c). The spacing between two peaks (Ga atoms) is ∼6.4 Å and agrees well with twice the GaN lattice constant, 3.189 Å.

Also, the spacing between the two adjacent bright rows (indicated by white arrows) reveals to be 11.4 ± 0.6 Å. It agrees well with 11.05 Å, 4 times the inter-planar spacing of the GaN and observed from RHEED analysis of the quantum height islands as weak 4× streaks. It is also the consistent spacing between bright (or dark) rows in both islands despite the distinct surface morphology. This 4× periodicity corresponds to the spacing between two dashed lines drawn through the surface models in Fig. 4. It should be emphasized that the model shown in Fig. 4 (e), corresponding to the 5-layer island and the STM image shown in the Fig. 4 (d), is still anisotropically strained along $[1\bar{1}0]$ and $[11\bar{2}]$.

However, the zoom-in STM image on the 6-layer island surface as presented in Fig. 4 (g) reveals distinct morphology and the different insight for its lattice. The line section B given in Fig. 4 (c) is a measurement along a Mn-Ga row, as drawn with a black line in Fig. 4 (g). The spacing between two peaks (Ga atoms) is measured to be 5.6 ± 0.3 Å, which is smaller than that for the 5-layer island (∼6.4 Å) and agrees well with twice the δ-phase MnGa lattice constant (2.76 Å). It can be implied that the MnGa lattices on the 6-layer island surface relax back to the primitive δ-phase MnGa (111) lattices with a unit cell as marked with black lines in Fig. 4 (h). In consequence, there is no strain at least along $[1\bar{1}0]$. In fact, the relaxation process on the 6-layer island surface does not take place as a whole. While the peak spacing along $[1\bar{1}0]$ shrinks back to the original value for δ-phase MnGa, the spacing between two adjacent bright rows is measured to be 10.8 ± 0.4 Å and remains the 4× periodicity as on the 5-layer island. Therefore, it can be deduced that one part shrinks while the rest expands along $[11\bar{2}]$ within the 4× periodicity.

Based on these observations, the surface model of the 6-layer island surface is derived and presented in Fig. 4 (h). Two unit cells conjugate within the 4× periodicity as illustrated in Fig. 4 (h), each marked with black and green lines. The black unit cell is obtained from the 13% shrinkage of the unit cell shown in Fig. 4 (e) along $[1\bar{1}0]$ and $[11\bar{2}]$, while the green one is from the 13% shrinkage of the same unit cell along the MnGa $[1\bar{1}0]$ and additional 13% expansion along $[11\bar{2}]$. The black unit cell is the one which relaxes and closely matches the primitive δ-phase MnGa (111) lattices, so that it would relieve the strain induced by the lateral expansion and result in 4% of strain along $[11\bar{2}]$. In contrast, the green unit cell is the one relaxing only along the $[1\bar{1}0]$ but resulting in more strain, as much as 37% along $[11\bar{2}]$.

Overall, the 4× periodicity along $[11\bar{2}]$ keeps through the relaxation process. Overlaying

Fig. 4 (h) on the zoom-in STM image as presented in Fig. 4 (g), it is found that the Ga atoms (black spheres) are superimposed exactly on the bright atomic features along the zig-zag row structures represented by the black unit cell. In fact, it resembles the simulated and experimental STM images for the 2×2 reconstruction surface of the δ-phase MnGa (111) given in ref. [13]. The still-strained lattices represented by the green unit cell appear as dark valleys in the STM image.

Comparison of the line sections C and D shown in Fig. 4 (f) can give profound insights regarding the distinct surface morphology of the islands. The line section C is a measurement across two bright rows diagonally on the 5-layer island surface, as marked with a blue line in Fig. 4 (d) and the line section D on the 6-layer island surface, as marked with a green line in Fig. 4 (g). In the line section C, the spacing between two high peaks is twice the spacing from the line section A. The low peak is deemed to be half way between two high peaks, therefore, the spacings between peaks (Ga atoms) are the same either in the line section A or C, as expected from the unit cell shown in Fig. 4 (e). The low peak corresponds to the relatively bright atomic feature along the dark Mn-Ga row (indicated by a black arrow), which corresponds to a Ga atom protruding less than Ga atoms along bright Mn-Ga rows (indicated by white arrows). In contrast, the line section D consists of split peaks, corresponding to the two Ga atoms on each bright row, corresponding to the zig-zag pattern with the larger STM corrugation.

We therefore find that partial strain relaxation plays a role in this QSE-based system MnGa/GaN. Although electronic growth per se does not require invoking the traditional concept of a critical thickness and strain relaxation, there is no reason why these concepts should not also continue to apply in QSE systems such as this. In this case, the built-up strain within the 5-layer islands is partially relaxed in the 6-layer islands, but still the 6-layer height islands only are stable, and we do not find 7- or 8-layer height islands. Therefore, it further shows that strain relaxation by itself cannot account for the existence of quantum height island structures.

IV. CONCLUSIONS

We have studied the spontaneous formation of MnGa(111) into two distinct quantum height islands, the 5-layer and the 6-layer islands, which have grown onto the Ga-rich N-polar

GaN($000\bar{1}$) surface in pseudomorphic growth mode. We have presented atomic resolution STM images providing a direct insight into surface models and proposed the possible surface models. The 5-layer island surface has 2×2 reconstructed δ-phase MnGa (111) lattices under considerable strain due to the in-plane expansion (15.6-20.5%) which is accompanied with the out-of plane contraction (-13.6 %). On the 6-layer island surface the strain is partially relieved through relaxation to the primitive δ-phase MnGa (111) lattices along the bright rows but still remains along the dark valleys. We see that this non-uniform alternating strain can allow more energetically favorable row structures on the 6-layer island surface. It can explain the frequent and consistent observation of the 5-layer islands being less stable and transitioned into 6-layer islands even during a STM measurement.[8] Finally, we conclude that the existence of, or the need for, strain relaxation (or even partial relaxation) does not interfere with the existence of quantum height island structures, nor is strain relaxation by itself sufficient to account for their existence.

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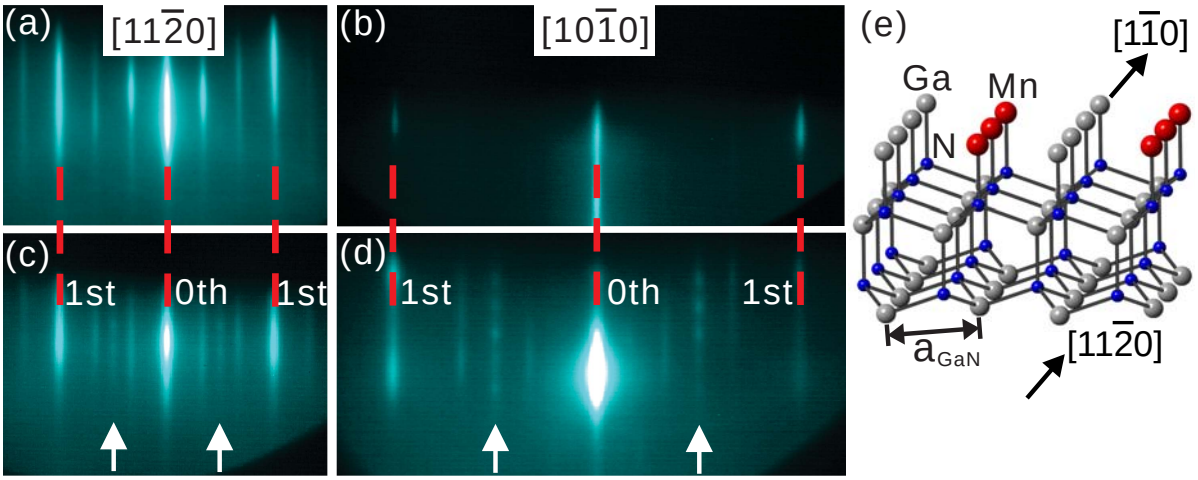

FIG. 1: (a), (b) RHEED patterns of GaN(000$\bar{1}$) substrate before the MnGa island formation along [11$\bar{2}$0] and [10$\bar{1}$0] at ∼250°C; (c), (d) RHEED patterns after the MnGa island formation. Red dashed lines drawn along the primary streaks, and arrows pointing out the additional half order streaks. (e) Perspective view of the interface of MnGa island and GaN substrate.

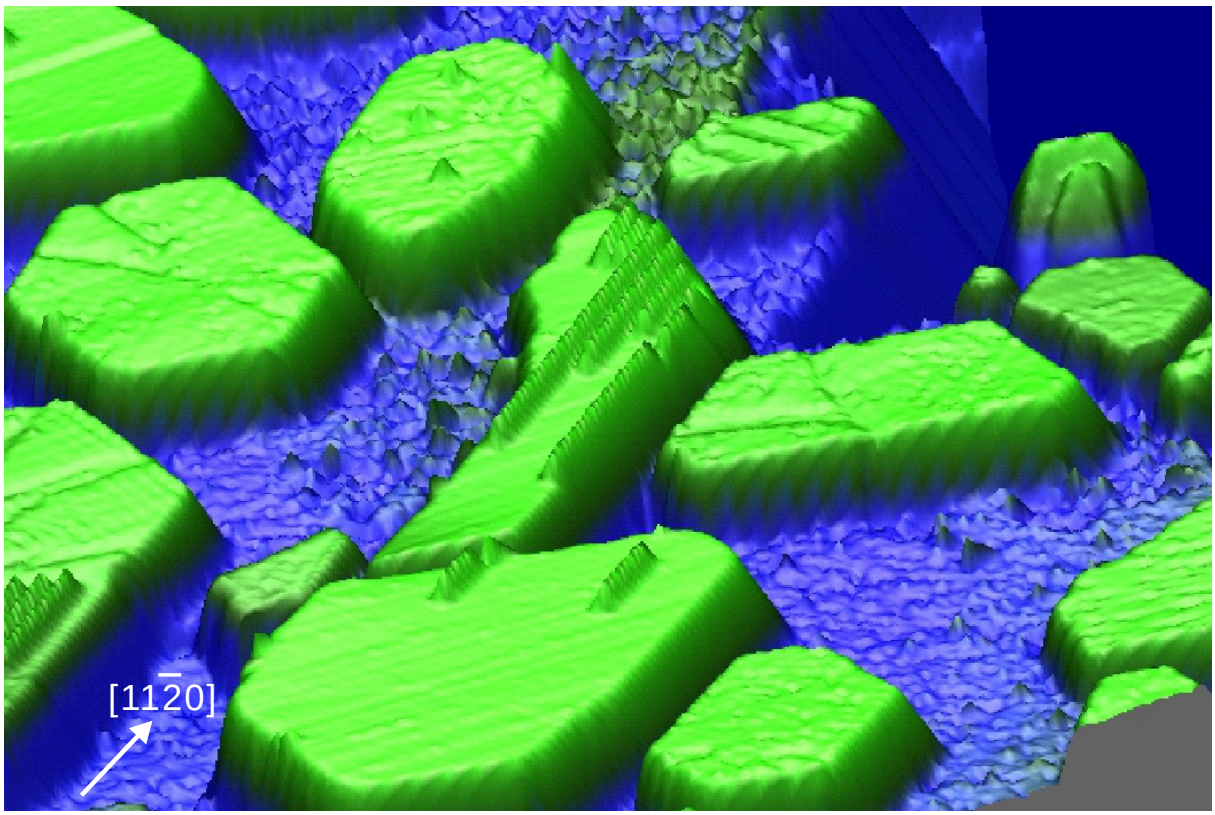

FIG. 2: 200 nm × 120 nm 3D-rendered STM image of MnGa islands on N-polar GaN(000$\bar{1}$) surface. Scanning parameters: V_S = -2.44 V and I_T = 103 pA.

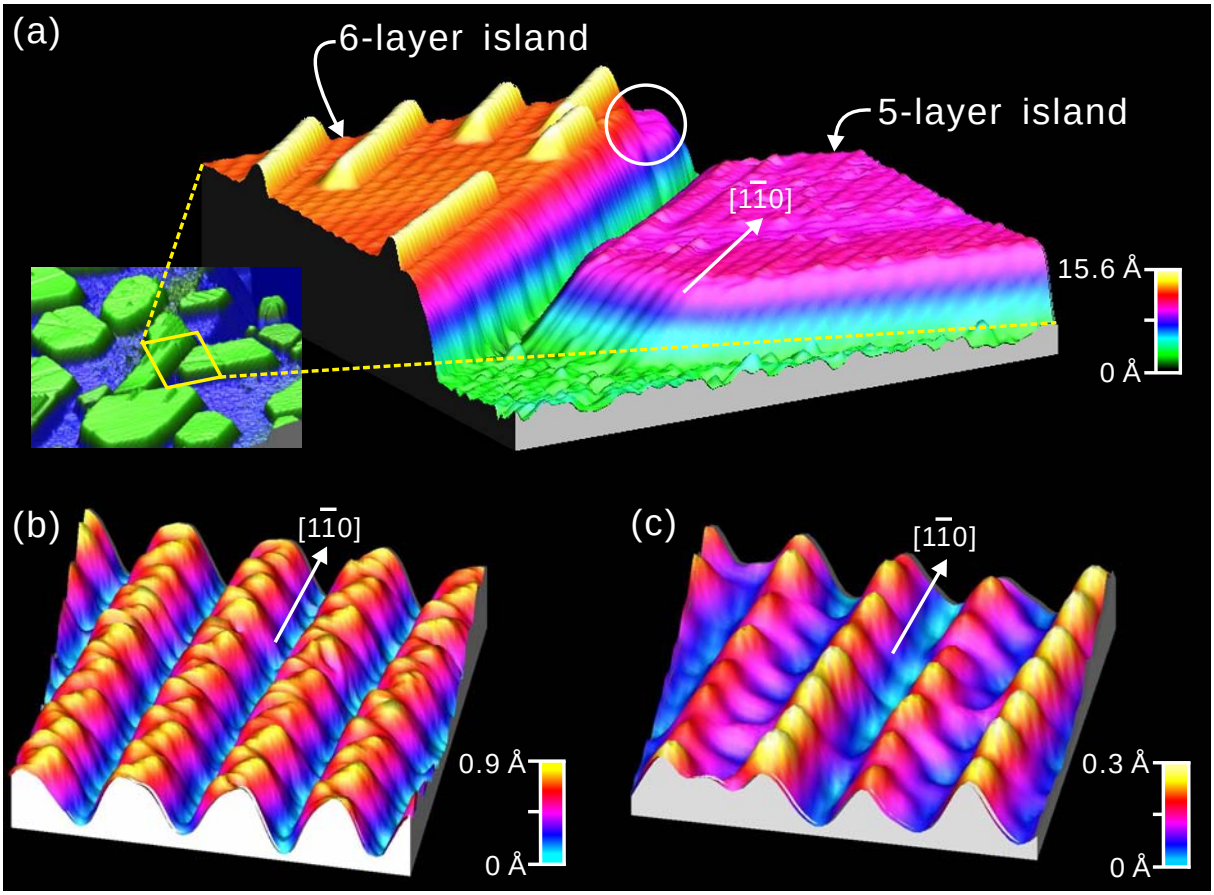

FIG. 3: 3D-rendered STM images of MnGa islands on N-polar GaN(000$\bar{1}$) surface. (a) 40 nm × 40 nm (V_S = -0.87 V and I_T = 155 pA); (b) 4 nm × 4 nm of 6-layer MnGa islands (V_S = -1.2 V and I_T = 80 pA); (c) 4 nm × 4 nm of 5-layer MnGa islands (V_S = -0.91 V and I_T = 153 pA).

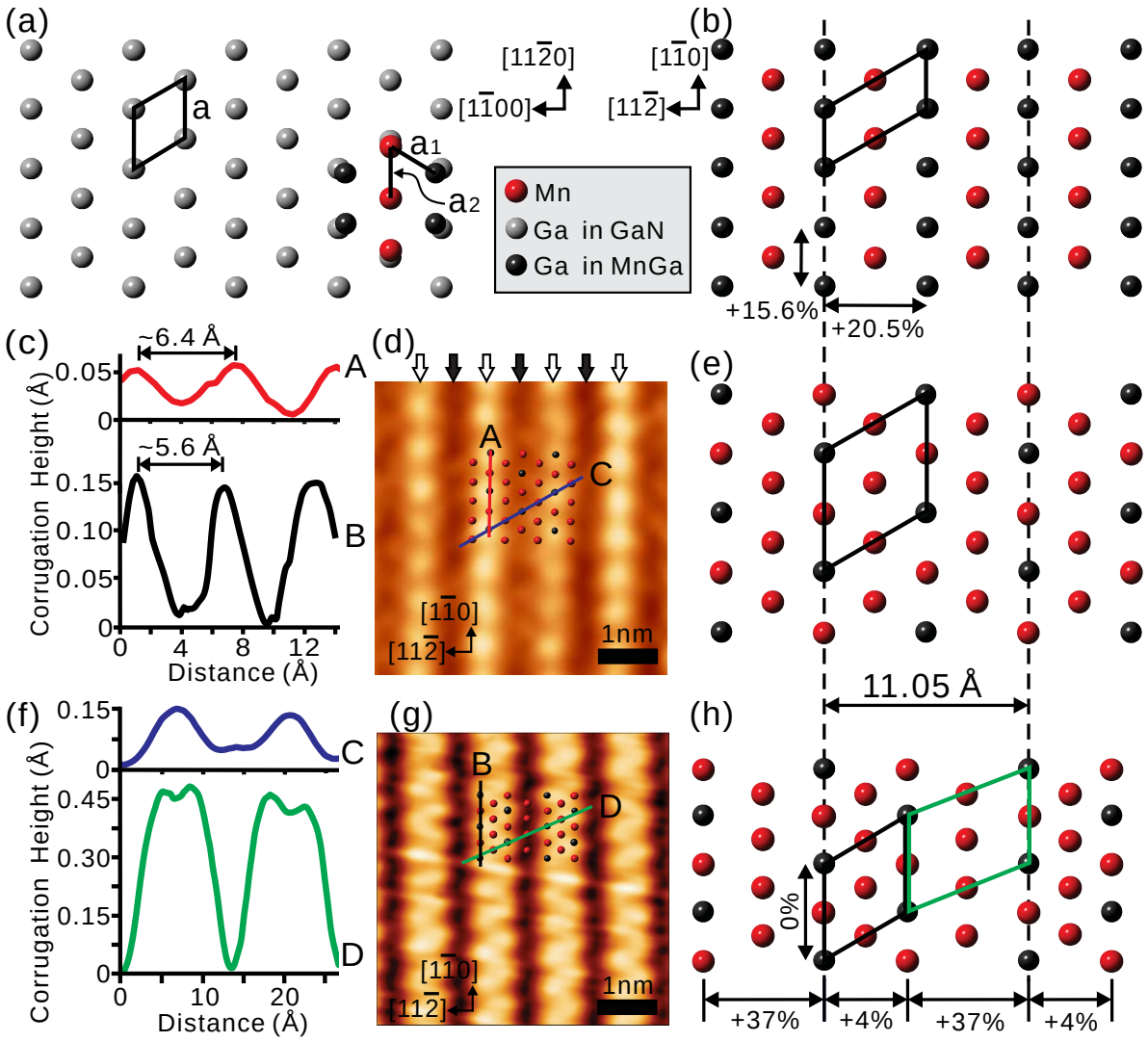

FIG. 4: (a) Ga adlayer lattice of N-polar GaN with a 1 × 1 unit cell (a = 3.189 Å) and primitive δ-phase MnGa (111) lattice (a_1 = 2.67 Å and a_2 = 2.76 Å). Ga atoms in GaN and MnGa illustrated in gray and black, respectively. (b) Surface model of anisotropically strained MnGa(111) having a 1 × 2 unit cell. (c) Line profiles along a row structure on the 5-layer island (A) and the 6-layer island (B). (d) 5 nm × 5 nm zoom-in STM image of the 5-layer island surface showing its surface model given in (e) and the line profile paths. White and black arrows indicating bright and dark Mn-Ga rows, respectively (V_S = -0.91 V and I_T = 153 pA). (e) Surface model of reconstructed, anisotropically strained MnGa(111) having a 2 × 2 unit cell for the 5-layer island. (f) Line profiles diagonally across two row structures on the 5-layer island (C) and the 6-layer island (D). (g) 5 nm × 5 nm zoom-in STM image of the 6-layer island surface showing its surface model given in (h) and the line profile paths (V_S = -1.2 V; I_T = 80 pA). (h) Surface model of MnGa(111) for the 6-layer island having non-uniform alternating strain.