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Misfit relaxation of InN quantum dots: Effect of the GaN capping layer

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The strain state on InN quantum dots (QDs) over GaN/sapphire substrates was analyzed by transmission electron microscopy. Changes in the in-plane lattice parameter of uncapped and capped InN QD heterostructures have been measured using moiré fringe analysis. The uncapped QDs are almost completely relaxed, due to a misfit dislocation network present at the InN/GaN interface without generating any threading dislocations inside the QDs. In addition, a low-temperature-GaN capping process on InN QDs heterostructures was evaluated. Although this deposition avoids the InN decomposition, it modifies the QDs' morphology, decreases both the aspect ratio and, consequently, the plastic relaxation of the heterostructure.
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Recently, group III-nitrides have shown great promise in optoelectronic devices with a wide range of applications. In these devices, InGaN/GaN quantum wells constitute the active structure in light-emitting diodes and laser diodes, and can cover a wide spectral range from near ultraviolet to red. In particular, InN is of great interest due to the possibilities it offers for devices operating at potentially infrared frequencies.[1] Its small band gap ($\sim$0.7 eV) (Ref. 2) should be extremely useful for telecommunication-wavelength devices. The combination of the inherent properties of InN and self-assembled nanostructures with quantum confinement presents further possibilities for this material. Moreover, quantum dot (QD) structures are especially promising because they act as electron-hole recombination centers, increasing the emission efficiency and potentially acting as single-photon emitters.[3] However, high-quality InN deposition is still not well established due to the complexity required for good epitaxial growth. The body of work on InN heteroepitaxy using transmission electron microscopy remains sparse.[4–6]

In mismatched heteroepiaxial growth: it is well-known that strain can be accommodated elastically or plastically depending on the lattice mismatch and the surface energy of the materials involved.[7] The strain relaxation mechanism depends not only on the lattice mismatch but also on the growth conditions; in the present case, the substrate temperature and metal/N ratio.[8,9] Three-dimensional (3D) growth of InN on GaN (system with a 10% compressive lattice mismatch) via the Stranski-Krastanov mode has been previously reported.[10,11] Nevertheless, it seems that even if the growth of InN on GaN is 3D, the strain in epitaxial InN is initially relieved by dislocations rather than surface islanding.[11] While growth of coherent InAs islands on GaAs is very well known,[12] there are only a few studies of strain relaxation of individual InN islands and the relationship to their size or shape.[13]

Here, we have used the moiré fringes seen in plan-view transmission electron microscopy (PVTEM) images to determine the strain relaxation of individual InN QDs. We show that a misfit dislocations (MD) network at the InN/GaN interface accommodates the majority of the misfit strain. In addition, we also analyze the influence of a low-temperature (LT)-GaN capping process on the InN QDs. The direct growth of GaN is problematic since the optimum growth temperature for GaN is 1050 °C and InN starts to decompose at 650 °C.[14] We will show that this process avoids the re-evaporation of InN QDs, although it changes the QD shape (diameter, height, and aspect ratio) as well as the residual strain.

Two InN QD samples were grown on GaN/Al_2O_3 by metalorganic vapor phase epitaxy, depositing in the second a LT-GaN capping layer. First, a buffer layer of GaN was grown on (0001) sapphire using the usual two-step process[15] at a temperature close to 1000 °C. The temperature was then lowered to 550 °C and InN QDs were deposited using a V/III ratio of 15000 and NH_3 as a nitrogen precursor. In order to solve the InN decomposition, we used a double step process where GaN is first deposited at low temperature (550 °C) above the InN dots, in order to cover the dots and to prevent their decomposition at a higher temperature. Once protected in such a way, the growth temperature is raised to 1050 °C, to recrystallize the low-temperature GaN previously deposited. This is necessary because the low-temperature GaNhas poor crystallinity. The uncapped and GaN-capped samples are denoted U1 and C1 here. Conventional and high resolution transmission electron microscopy (TEM) (HREM) in cross section (XTEM) and PVTEM were carried out in a JEOL 2011 and a JEOL 2010 FEG, both working at 200 kV.

In a previous study,[16] using resolved moiré fringes in PVTEM specimens,[17] we showed that InN QDs nucleated where a threading dislocation (TD) intersected the surface of the GaN layer. The TD did not propagate into the QD. It is possible to determine the strain in the QD by a more detailed analysis of the moiré fringes,[4] and we apply this technique

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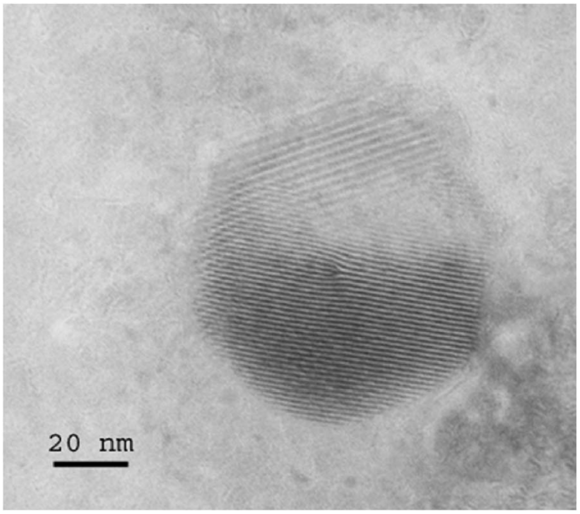

FIG. 1. Micrograph in planar view orientation, showing moiré fringes pattern, of the uncapped heterostructure (sample U1).

here to determine the crystalline relationship between the GaN and QDs with and without the capping layer. A PVTEM bright-field image of sample U1, with only one set of $\mathbf{g}=1\bar{1}00$ in the InN/GaN system, is shown in Fig. 1(a). A well-defined translational moiré fringe pattern is observed. The spacing of translational moiré fringes is

$$D_m = \frac{d_e^{\mathrm{QD}} d_s}{d_e^{\mathrm{QD}} - d_s}, \quad (1)$$

where d_e^{QD} and d_s correspond to the $\{1\bar{1}00\}$ plane spacing in the QD and GaN buffer layer, respectively. In the present case $d_s=0.2762$ nm and, from Eq. (1), an estimate of the plastic relaxation, δ, in the heterostructure is

$$\delta = \left(1 - \frac{\epsilon_r}{f}\right) = \frac{d_s - d_e^{\mathrm{QD}}}{d_s - d_{\mathrm{InN}}}, \quad (2)$$

where ϵ_r is the residual strain, f is the lattice misfit, and $d_{\mathrm{InN}}=0.3059$ nm corresponds to the $\{1\bar{1}00\}$ spacing planes in fully relaxed InN. In order to obtain an accurate strain relaxation value, several InN QDs were analyzed, giving an average moiré fringes spacing of $D_m=2.9\pm0.2$ nm. Applying Eq. (2) gives a plastic relaxation of $\delta=97\pm6\%$.

To confirm this result, HREM images were recorded along the $\langle 0001 \rangle$ zone axis in areas containing InN QDs, as shown in Fig. 2(a). A magnified region of this image is shown in Fig. 2(b) which contains interruptions in the lattice fringes due to the MDs. The MDs are introduced in a successive sequence of atomic $\{1\bar{1}00\}$ planes as can be more easily observed in a Fourier filtered image [inset in Fig. 2(b)]. Extra half-planes in the lattice fringes are indicated by white arrows. On average, an extra half-plane can be observed every 10.5 $\{1\bar{1}00\}$ GaN planes or equivalently 9.5 $\{1\bar{1}00\}$ InN planes, which gives a plastic relaxation of 96%, very close to the previously determined applying Eq. (2). Thus, the InN QD is not completely relaxed (but very close to it) in the analyzed area since complete relaxation requires a MD every 10.3 $\{1\bar{1}00\}$ GaN planes. This demonstrates that the compressive strain in the InN QDs grown on GaN is mainly relaxed through a MD network at the InN/GaN interface and, consequently, the percentage of strain relief by surface islanding has to be very low. Therefore, the mechanism of InN QDs formation has to differ notably with respect to the Stranski-Krastanov mechanism, observed in other heteroepiaxial systems,[18] being more similar to the Vollmer-Weber growth. Remarkably, despite the high MDs' density at the interface, TDs are not observed inside the InN QDs (Ref. 16) being an interesting result for the fabrication of InN QDs-based optoelectronic devices.

Morphological parameters, such as average QD diameter and height as well as dot density, were measured in samples U1 and C1 to determine the effect of the capping layer. The measured InN QDs density was the same in both samples (4×10^8 cm^{-2}), as may be expected since the only difference in growth was the LT-GaN capping layer in sample C1, i.e., after InN dot nucleation and growth. The InN QDs in sample U1 are flat regular hexagonal prisms, with an average diameter (w) and height (h) of 73 ± 11 nm and 12 ± 2 nm, respectively (XTEM measurement),[16] i.e., the aspect ratio h/w is about 1/6. The LT-GaN capping layer gives rise to clear shape changes in the InN QDs. The change in the InN QD diameter can be seen by comparing Figs. 3 and 1. After capping, the average QD diameter has increased to 110 ± 30 nm, while the height has decreased to 6 ± 2 nm. Therefore, although the QDs density remains constant, the aspect ratio decreases considerably ($h/w=1/18$). Consequently, the flat hexagonal shape QDs present a morphological instability during the LT-GaN capping layer growth, which suggests that the original U1 aspect ratio is far from equilibrium at the C1 growth conditions. The C1 heterostructure is grown by a two-step procedure with growth temperature and N_2 flow changes involved. The thermal instability due the low InN dissociation temperature and high equilibrium N_2 vapor pressure over the InN (Ref. 14) would imply nitrogen desorption

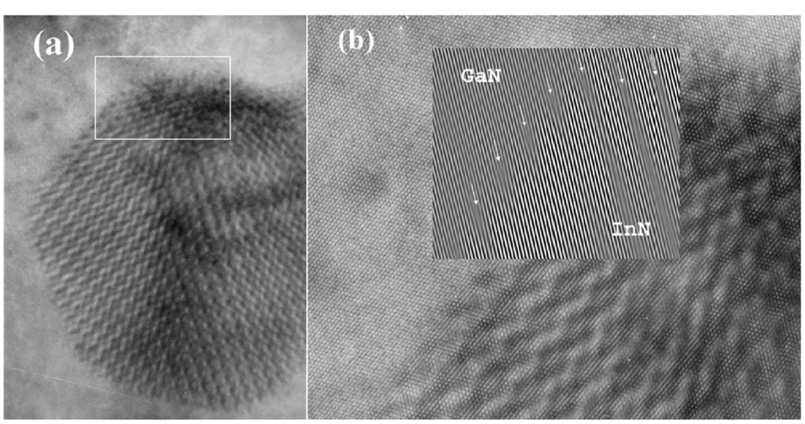

FIG. 2. (a) Plan-view HREM image along the [0001] axis of InN/GaN. (b) Magnified region corresponding to the rectangle area in (a). The inset micrograph is a result of a Fourier filtering treatment of the contrasts. The interruptions in the lattice fringes are indicated by white arrows.

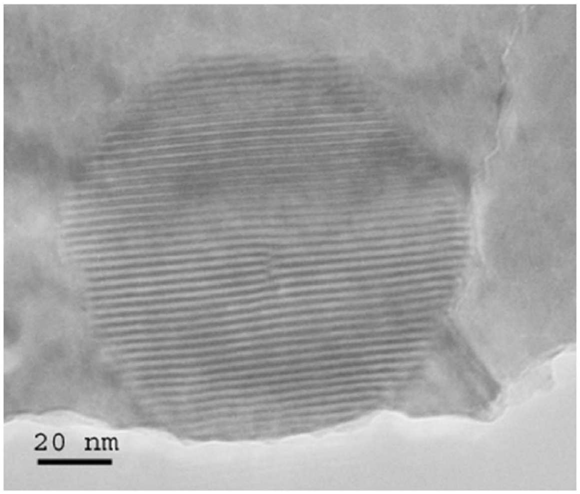

FIG. 3. PVTEM micrograph of the LT-GaN capped sample (C1) where the moiré fringes pattern can be clearly observed.

from dots undergoing surface changes.[19] Not only does the InN QD shape change with the capping layer; the strain relaxation of the heterostructure also varies. The strain relaxation in sample C1 was also determined by analyzing moiré fringes in several InN QDs. The LT-GaN capping layer gives rise to an increase of the average fringe spacing, $D_m = 3.2 \pm 0.2$ nm. Applying Eqs. (1) and (2) to sample C1, this indicates that plastic relaxation decreases to $85 \pm 5\%$. This decrease of the plastic relaxation degree can be explained by the reduction of the QD height according the classical model of Matthews.[20] The LT-GaN capping layer incorporation in the heterostructure modifies the MD spacing. The evident high mobility of the QD constituting elements, able to modify the QD aspect, should be the main reason for the MD rearrangement. In any case, the two-step growth procedure supposes an improvement for the development of InN QD-based devices. Further research is in progress to understand and control the InN QDs shape instability, as it represents a key point for these heterostructures.

In summary, the uncapped InN/GaN QD heterostructure is almost completely relaxed $(97 \pm 6\%)$ by the formation of MDs in the interface area, and a very low percentage of strain is relieved by surface islanding. Therefore, the mechanism of InN QDs formation has to differ notably with respect to the Stranski-Krastanov mechanism, observed in other heteroepiaxial systems. Additionally, TDs have not been observed in the InN QDs. To overcome the problem of InN thermal decomposition, a two-step LT-GaN capping process is evaluated. The incorporation of this LT-capping layer avoids this effect, although it modifies the InN QDs morphological parameters with a decrease in both aspect ratio and plastic relaxation degree. The control of the shape instability during the capping layer growth supposes a crucial step for the advance of InN QD devices.

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