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Doping modulation in GaN imaged by cross-sectional scanning tunneling microscopy

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We investigated the imaging mechanisms of a Si doping modulation in GaN by cross-sectional scanning tunneling microscopy (STM). The Si doping modulation gives rise to a voltage and tip dependent height modulation of at least 0.4 Å. The origin of the height modulation in constant-current STM images is traced to two mechanisms. A doping-induced modulation of the band edge energies yields a voltage dependent electronic contrast and an additional mechanical relaxation of the doping-induced strain at the cleavage surface is responsible for a voltage independent modulation of 0.35 Å. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3123258]

Epitaxial layers of group-III nitrides developed rapidly toward the system of choice for green to ultraviolet optoelectronic devices. Such devices are based primarily on heterostructures of different group-III nitrides and/or differently doped layers,[1] where the exact atomic arrangement within the epitaxial layers of the nanostructures and at the interfaces sensitively influences the optoelectronic properties.[2] A particularly attractive tool for the high resolution analysis of interfaces and epitaxial layers in compound semiconductors is cross-sectional scanning tunneling microscopy (STM).[3,4] Indeed, for zincblende type materials, cross-sectional STM provides a direct atomically resolved access to *structural and electronic* bulk properties,[4,5] thereby helping to unravel the processes during epitaxy. This technique is in principle also applicable to wurtzite structure semiconductors due to the presence of suitable cleavage surfaces.[6] However, up to now, no epitaxial structure or interfaces in group III-nitrides could be successfully imaged by cross-sectional STM. Here we investigated a doping modulation in GaN by cross-sectional STM and identified the contrast mechanisms. We demonstrate that the doping modulation in GaN is imaged through both a dopant-induced variation of the band edge energies and a dopant-induced strain relaxation at the cleavage surface.

For our experiments we used epitaxial GaN layers with a *n*-type Si doping modulation grown on the *c* plane. They were cleaved in ultrahigh vacuum ($<10^{-8}$ Pa) along a $(1\bar{1}00)$ plane, opening a cross-sectional view on the epitaxial layers. For the STM measurements, we used $Pt_{0.8}Ir_{0.2}$ tips and acquired the data using a RHK SPM control unit.

Figure 1 shows an overview of the GaN $(1\bar{1}00)$ cleavage surface. The constant-current STM image was measured at positive sample voltage, thus imaging the empty density of states corresponding to the empty dangling bonds localized above the Ga surface atoms forming atomic rows along the $[11\bar{2}0]$ direction.[6] The surface consists of atomically flat terraces separated by mono- and diatomic steps. These steps are oriented primarily along the [0001] direction and are mostly cleavage related. Only few steps originate from dislocations.[7]

In addition, Fig. 1 shows a contrast modulation along the [0001] direction with about six periods visible in this particular image. The maxima of the contrast modulation are marked by arrows. This contrast modulation is not influenced by any surface defect, e.g., steps. It is found to be independent of the step density. The average separation between adjacent maxima of this contrast modulation is 270 ± 30 nm.

In order to analyze the contrast modulation, we extracted height profiles along the [0001] direction from an atomically flat terrace [Fig. 2(a)]. All height profiles acquired for different voltages *using the same tip* (labeled tip 1) exhibit very similar height modulations. However, at a close look, the modulation amplitude is decreasing slightly with the applied voltage, as illustrated by the corresponding root mean square (rms) roughness as a function of voltage in the inset of Fig.

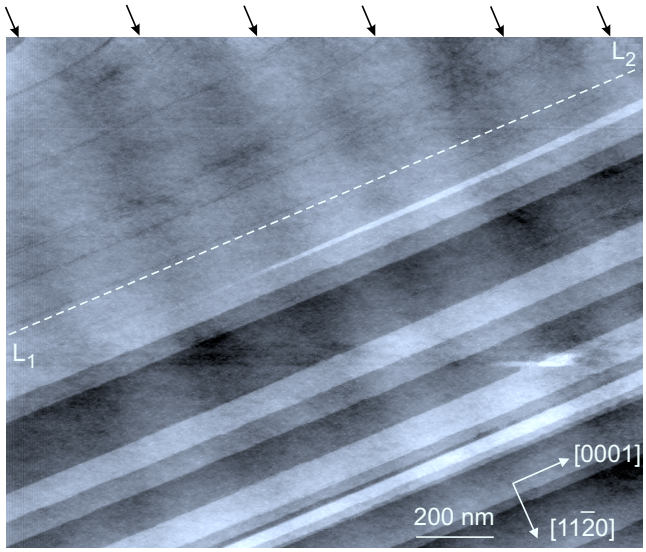

FIG. 1. Cross-sectional empty state constant-current STM image showing a doping-induced contrast modulation along the [0001] direction in GaN. The maxima are marked by arrows. The steps, oriented primarily along the [0001] direction, do not affect the contrast modulation. The image was measured at +5.9 V applied to the sample and 80 pA tunnel current.

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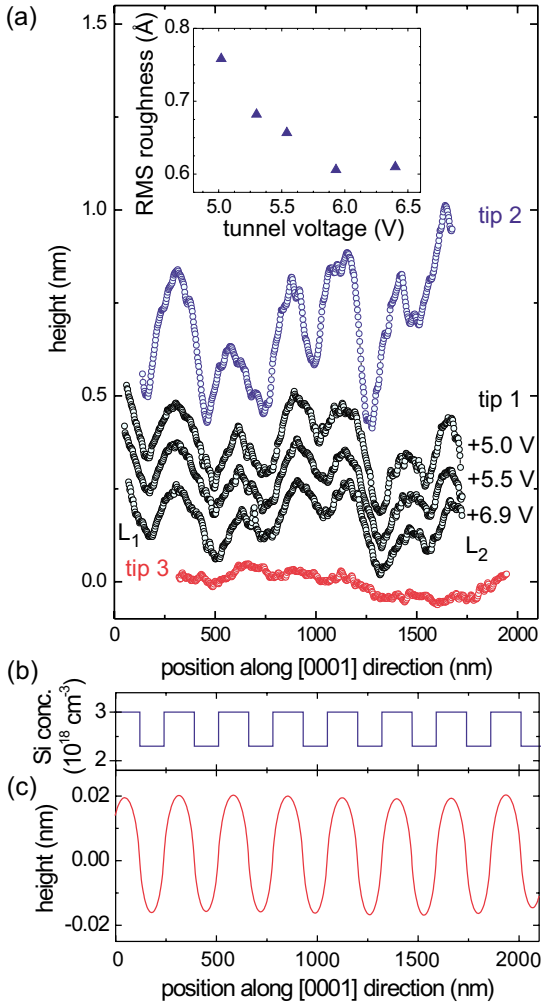

FIG. 2. (Color online) (a) Height profiles of the contrast modulation along the dashed line in Fig. 1. The profiles were acquired at different voltages and with three different tip configurations. The profiles are offset for clarity. Inset: rms roughness of the height profiles for different tunneling voltages acquired with tip 1. (b) Si concentration profile used to simulate the height modulation in (c). (c) Calculated height corrugation arising from the relaxation at the GaN cleavage surface of the strain induced by the model Si doping modulation shown in (b).

2(a). In contrast, *for different tip configurations*, obtained by tip instabilities during repeated scanning of the same location, the amplitude is changing drastically. For example, tip 2 yields a very large modulation height (corresponding rms roughness is 1.4 Å), whereas tip 3 yields a very small modulation height with a corresponding rms roughness of only 0.3 Å. In the latter case the modulation peak to peak height of ≈0.4 Å is very close to the noise level, which thus dominates the rms roughness. We observed a contrast modulation with all tips. The smallest observed modulation height was found to be close to 0.4 Å, as shown, e.g., for tip 3. Note that the positions of the modulation maxima and minima do not shift with different tunneling voltages or tips.

In order to discuss the origin of the observed contrast modulation, we turn to the Si concentration measured by secondary ion mass spectroscopy (SIMS) [Fig. 3]. The Si doping concentration varies between 2.3×10^{18} and 3.0×10^{18} cm^{-3}. The maxima of the modulation of the Si concentration have an average separation of 270 ± 20 nm. For other epitaxial GaN layers (not shown here) without doping modulation, we did not observe any comparable contrast

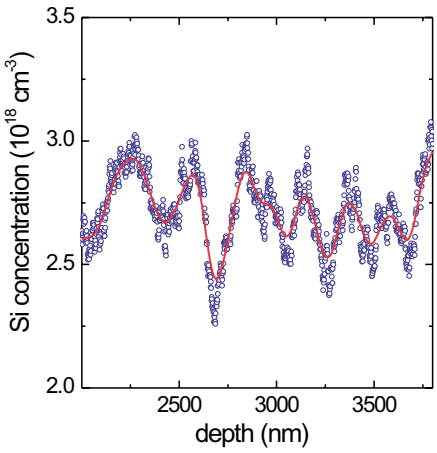

FIG. 3. (Color online) Si concentration along the [0001] direction measured by SIMS (circles). The solid line is a smooth for guiding the eye.

modulation. Furthermore, the separation of the maxima of the Si concentration agrees well with the distance between neighboring maxima of the contrast modulation. This indicates that the contrast modulation arises from the Si-doping modulation.

One might think also of a different origin of the contrast modulation; standing electron waves. However, the observed wavelength of 270 nm would be extremely long as compared to previous observations of standing waves on, e.g., Cu(111) (Ref. 8) or GaAs(110) (Ref. 9) surfaces. Such a long wavelength would correspond to extraordinary low effective masses. Furthermore, surface steps would act as scattering obstacles and thus reflect standing waves. As a result, their wave orientation would be perpendicular to the steps, in contrast to our observation of no step correlation (see Fig. 1). Thus, standing electron waves can be excluded as origin of the contrast modulation.

Thus, we turn to the Si doping. The change of the Si concentration shifts the energy position of the conduction band edge E_C relative to the Fermi energy E_F. Thereby, the Si doping concentration increases the number of states on the GaN surface, into which electrons can tunnel at fixed voltage. This increased current in higher doped layers, is compensated by the feedback loop in the constant-current mode by increasing the tip-sample separation. This leads to the measured height modulation in the constant-current STM images in analogy to that of other local band edge changes.[10]

The contrast of differently doped areas is also voltage dependent: The number of states, into which electron can tunnel, increases with the voltage V, because all states between E_F and E_F+eV contribute to the tunnel current. Thus, the total current increases with voltage but the current into the additional states made available for tunneling because of a local Si doping increase (due to modulation doping) remains essentially unchanged. This leads to a decreasing contrast modulation with increasing voltage in agreement with the observation in the inset of Fig. 2(a). Similar effects have been observed also on other III-V semiconductor surfaces around charged dopant atoms and defects (screened Coulomb potential-induced band edge shift)[11] and at InAs quantum dots in GaAs (additional quantum dot states).[12]

The strong effect of different tips on the magnitude of the modulation can be explained by taking the additional tip-induced band bending into account. The presence of the

biased tip induces a strong electric field, which cannot be screened directly at the semiconductor surface, due to its limited free carrier concentration. Therefore, the electric field of the tip penetrates into the semiconductor and induces a tip-induced band bending.[13] This tip-induced band bending leads to a shift of the band edges at the surface relative to those in the bulk (and to E_F). Thus, the number of states available for the tunneling electrons decreases with the band bending. Thereby the relative fraction of the locally increased doping-induced number of states with respect to the total current and the resulting modulation contrast, increase again. The band bending and thus the modulation is largest for a wide, blunt tip, for which the tip-induced band bending can be approximated using the one-dimensional model.[13] For an atomically sharp tip, in contrast, the electric field is strongly localized. This leads to a significantly reduced band bending[14] and therewith to a reduced modulation. Furthermore, the tip-induced band bending is doping dependent, which leads to an additional modulation contrast. Thus, the effect of the tip-induced band bending depending on the sharpness of the tip can explain the strong tip sensitivity of the modulation height observed in Fig. 2(a). This also demonstrates that the surface is unpinned (i.e., no intrinsic surface states are in the fundamental band gap) in agreement with Ref. 6.

We recall that that the contrast modulation never disappears completely even at very large voltages and for sharp tips. The modulation height is always ≥ 0.4 Å. This nondisappearance at large voltages is in contrast to previous observations of, e.g., charged dopants and defects, where the extended height change induced by the screened Coulomb potential disappears at large voltages.[11] Therefore, the Si doping modulation also induces a mechanical bending of the surface, which is independent of the electronic parameters of the tunneling process.[12] Thereby a constant background modulation is induced, superimposed by electronic effects.

A mechanical bending of a surface typically originates from the strain relaxation upon cleavage.[12,15] In GaN strain can be introduced by Si doping because GaN layers with different Si doping concentrations were found to exhibit slightly different lattice constants.[16] On basis of these data we calculated the lattice constants (a in $[11\bar{2}0]$ and c in $[0001]$ direction) of the lower and higher Si doped layers present in our sample (see Fig. 3). Using Si doping concentrations of $n_l = 2.3 \times 10^{18}$ cm^{-3} and $n_h = 3.0 \times 10^{18}$ cm^{-3} as determined from SIMS, we obtained lattice constants of $a_l = 3.187\,084$ Å and $c_l = 5.185\,937$ Å as well as $a_h = 3.187\,880$ Å and $c_h = 5.185\,583$ Å, respectively. Note that while a increases, c decreases with the Si doping concentration and only the values $(a_l - a_h)/a_l$ and $(c_l - c_h)/c_l$ are relevant for the following calculation. A slab of 15 periods of alternating 120 nm thick low (n_l) and 150 nm thick high (n_h) Si-doped GaN layers along the [0001] direction [Fig. 2(b)] and the corresponding above derived lattice parameters for the two doping levels were used in finite difference method calculations based on continuum mechanics. For this calculation, we used the stiffness constants of Ref. 17. From the calculated strain relaxation we extracted the corrugation height at the $(1\bar{1}00)$ surface along the [0001] direction [Fig. 2(c)]. The Si doping modulation leads to a structural relaxation of 0.35 Å independent of the tunneling conditions and in addition to electronic effects. This is in good agreement with the minimum measured modulation height of 0.4 Å. Furthermore, both contrast mechanisms of the Si doping modulation, strain relaxation and band edge shift, lead to higher modulation heights with increasing Si doping concentration. Thus, they add up, explaining that the modulation does not spatially shift with different voltages or tips.

In conclusion, we imaged an epitaxially grown Si doping modulation structure in GaN by cross-sectional STM. The Si doping modulation gives rise to a height modulation in constant-current STM images. The origin of the height modulation is traced to two contrast mechanisms, an electronic modulation of the band edge energies yielding a voltage dependent corrugation and a mechanical relaxation at the surface of the doping-induced strain superimposing a voltage independent contrast modulation.

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