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HTML ABSTRACT + LINKS

Scanning probe investigation of surface charge and surface potential of GaN-based heterostructures

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Scanning Kelvin probe microscopy (SKPM) and electrostatic force microscopy (EFM) have been employed to measure the surface potentials and the surface charge densities of the Ga- and the N-face of a GaN lateral polarity heterostructure (LPH). The surface was subjected to an HCl surface treatment to address the role of adsorbed charge on polarization screening. It has been found that while the Ga-face surface appears to be unaffected by the surface treatment, the N-face surface exhibited an increase in adsorbed screening charge density ($1.6\pm0.5\times10^{10}$ cm^{-2}), and a reduction of 0.3 ± 0.1 V in the surface potential difference between the N- and Ga-face surfaces. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1869535]

Investigation of the polarization behavior of nitride thin films, bulk crystals and heterostructures is of considerable interest for determining how interfaces, defects and inversion domain boundaries affect device performance. Scanning probe microscopy (SPM) techniques, including electrostatic force microscopy (EFM),[1] scanning Kelvin probe microscopy (SKPM),[2,3] and piezoresponse force microscopy (PFM)[4] have been previously employed to perform high-resolution characterization of the local electronic properties of III-nitrides. Of critical importance is to understand how the surface potential relates to the surface charge and thus, to the local electronic structure of GaN, which can be realized by the combination of SKPM and EFM. Bridger *et al.* have previously investigated GaN by EFM and SKPM, and the results have been used to determine a surface state density of $9.4\pm0.5\times10^{10}$ cm^{-2}.[1] In prior studies from our group, the surfaces of a GaN-lateral polarity heterostructure (LPH) have been investigated using PFM, Raman scattering and photoelectron emission microscopy (PEEM).[4–6] In this study, SKPM and EFM have been employed to measure the relative surface potentials and surface charge densities of Ga- and N-face GaN. In order to address the role of adsorbed charge in polarization screening on GaN, the measurements are made before and after a wet chemical treatment that modifies the surface in a controlled way.

Assuming a similar electron affinity, SKPM measurements of Ga- and N-face GaN are expected to reveal a potential difference approximately equal to the band bending (or surface work function) differences between the polar faces. Alternatively, EFM of the polar surfaces should respond to the net surface charge density, which is equal to the sum of polarization charge and (internal and external) screening charge.

The (1 μm thick) GaN-based LPH film was grown on a sapphire substrate using plasma induced molecular beam epitaxy.[2,7] The boundary between the Ga- and N-face GaN regions results in an inversion domain boundary (IDB). At the polar surfaces of (0001)-oriented wurtzite GaN crystals (spontaneous polarization, $P_{SP}=-0.034$ C/m^2), a divergence in the spontaneous polarization induces a polarization bound surface charge with a density of 2.12×10^{13} cm^{-2}.[8]

The sign of the polarization induced charge at each surface is related to the orientation of the polarization, and therefore, to the polarity of the crystal.[9] For epitaxial layers of wurtzite GaN with Ga-face polarity, the bound surface charge is negative, whereas for N-face GaN, the bound surface charge is positive. From Raman scattering measurements of these samples, the free electron concentration was determined to be $N_d=4.1\times10^{17}$ cm^{-3} for the N-face region and 2.5×10^{17} cm^{-3} for the Ga-face region.[5] It is expected that internal charge (free carriers, charged defects) and external charge (adsorbed charge) will act to screen the bound polarization charge. Charged surface states can also contribute to screening and additionally affect band bending. In our calculations, we assume that the magnitude of the bound polarization charge is the same for each face and that the internal screening mechanism is equivalent for each face.

Generally, surface cleaning processes are developed in order to remove native oxides, organic contaminants, metallic impurities, adsorbed molecules, and residual species as a fundamental step for improving device quality. In this study, we employ a well-documented HCl surface treatment in order to change the surface in a reproducible way to explore the polarization screening mechanism in GaN. For the surface treatment the sample was first submerged sequentially in trichloroethylene, acetone, methanol, and deionized water ul-

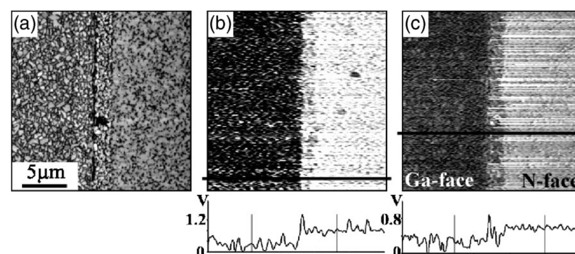

FIG. 1. (a) Topography of a 20×20 μm^2 area, (b) SKPM (with line profile) of a GaN-LPH prior to surface treatment, and (c) SKPM of the same area after surface treatment.

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trasonic baths for 10 min durations. The sample was then placed into HCl (38%) for an additional 10 min, before being rinsed for 3 min in deionized water and dried with N_2. It is expected that the surface will also have a significant amount of residual Cl, which has been reported to hinder reoxidation.[10]

A Park Scientific Instruments Autoprobe M5 AFM and rectangular Pt-coated Si cantilevers (5 N/m force constant, MikroMasch) were used in this study. In EFM, the force between a tip and surface is a combination of electrostatic and capacitive forces. The EFM image is constructed from the first harmonic (1ω) component of this force,[11]

$$F_{1\omega} = Q_t E_s + \frac{\partial C_t}{\partial z}(V_{\mathrm{dc}} - V_s)V_{\mathrm{ac}}, \quad (1)$$

where $Q_t = C_t V_{\mathrm{ac}}$ (first harmonic) is the charge on the tip, C_t is the capacitance of the tip–surface configuration, $E_s = \sigma/\varepsilon_0(1+\kappa)$ is the field due to an infinite sheet of uniform charge σ, $\kappa = 9.5$ is the dielectric constant of GaN, $\partial C_t/\partial z$ is the partial derivative of the tip–surface capacitance with respect to tip–surface separation, V_{dc} and V_{ac} (5.0 V_{rms} at 10 kHz) are dc and ac voltages applied to the tip, and V_s is the surface potential. The surface potential can be expressed as[1]

$$V_s = \frac{1}{e}\left(\phi_m - \chi_s - \Delta\chi_s - \Delta E_{fn} - \Delta\phi\right), \quad (2)$$

where ϕ_m is the metal workfunction of the tip coating, χ_s is the electron affinity of the surface, $\Delta\chi_s$ is the change in electron affinity due to a dipole effect, ΔE_{fn} is the position of the fermi level with respect to the bulk conduction band, and $\Delta\phi$ is the band bending.

In SKPM, the value of dc bias that minimizes the $F_{1\omega}$ signal [Eq. (1)] is equal to the surface potential (V_s), and by recording this value, an image of the surface potential can be constructed. The difference in surface charge density between the polar faces can be obtained from Eq. (1) and the dc bias that equalizes the force on the tip. Assuming that $\partial C_t/\partial z$ has the same magnitude but opposite sign for the polar faces we find:

$$|\sigma^{\mathrm{N}}| - |\sigma^{\mathrm{Ga}}| = \frac{\varepsilon_0(1+\kappa)}{C_t}\left(\frac{\partial C_t}{\partial z}\right)\left(2V'_{\mathrm{dc}} - V_s^{\mathrm{N}} - V_s^{\mathrm{Ga}}\right), \quad (3)$$

where V'_{dc} is the value of dc bias that equalizes the forces.

In measurements of the as-received sample, the SKPM revealed a surface potential of 0.3 V for the Ga face and 0.9 V for the N-face for a potential difference of 0.6 V as shown in Fig. 1(a) topography and Fig. 1(b) SKPM (with line profile), respectively. Following an HCl treatment, the surface potential did not change for the Ga-face and decreased to 0.6 V for the N-face [Fig. 1(c)]. The uncertainty in the measurements is estimated to be ±0.1 V, a value that takes into account reproducibility, noise, and variations in surface potential related to the sample roughness. Hsu *et al.* reported a 0.1 V reduction in surface contact potential for an HCl clean.[3] Cimalla *et al.* reported a potential decrease of ~0.1 V across an inversion domain boundary (IDB) (from N- to Ga-face side) in a GaN lateral polarity heterostructure (LPH) sample.[2] In our study, the measured potential difference is higher, but of the same order of magnitude. This difference could be due to variations in the sample or surface conditions.

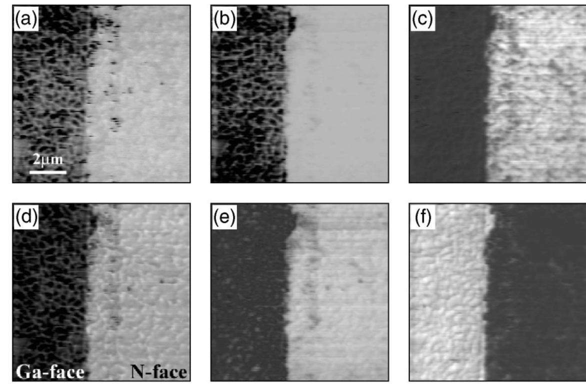

FIG. 2. (a)–(c) EFM phase and (d)–(f) EFM magnitude images of a 10 × 10 μm^2 region on the LPH-GaN sample with a dc bias of 0, 1, and 2 V, respectively.

The EFM $(V_{\mathrm{dc}} = 0)$ of the same area before the surface treatment revealed that the electrostatic force on the tip is larger for the N-face GaN. The EFM phase measurements indicated that the net surface charge (superposition of polarization and screening charge) is positive for the N-face surface and negative for the Ga-face surface. Following the surface treatment, the electrostatic force for the N-face further increased while the EFM phase measurements revealed that the net surface charge remained positive for the N-face surface and negative for the Ga-face surface.

In general, EFM results are difficult to quantify because the electrostatic force on the tip includes both Coulombic and capacitive components, therefore, we employ SKPM to measure the surface potential and deduce the net surface charge density by equalizing the electrostatic force on the tip for both polar surfaces. It was found that application of a dc bias could invert the EFM magnitude contrast of the two domains as shown in Fig. 2. Figures 2(a)–2(c) shows EFM phase and Figs. 2(d)–2(f) shows EFM magnitude images for tip biases of 0, 1, and 2 V, respectively, of the as-received surface. The results indicate that a tip bias of 1.5 V equalizes the electrostatic force on the tip from the Ga- and N-face regions, and the contrast reverses for a tip bias above 1.5±0.1 V. At 0 V bias, the tip responds to a net negative charge on the Ga-face GaN and a net positive charge on the N-face GaN. At this bias, the magnitude of the EFM indicates that the net surface charge on the N-face is greater, suggesting that the screening charge (external and internal) is greater for the Ga face. If we assume both faces have roughly the same degree of internal screening, the results suggest the Ga-face surface has more adsorbed charge. As the bias is increased, the second term in Eq. (1) is reduced for the N-face but increased for the Ga-face, which explains the change in magnitude contrast. This is demonstrated graphically in Fig. 3.

After the surface treatment, it was found that a tip bias of 2.0±0.2 V equalized the electrostatic force on the tip from the Ga- and N-face regions. While care was taken to perform this measurement as soon as possible after the surface treatment, it should be noted that this value varied on the time scale of several scans (~15 min), hence the larger uncertainty. We have determined the bias that equalizes the electrostatic force on the tip (both before and after the surface treatment) and can now employ this value to calculate the surface charge.

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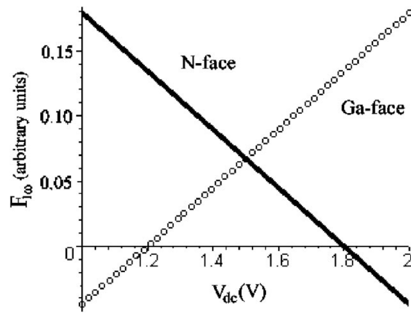

FIG. 3. Plot of electrostatic force as a function of applied dc bias demonstrating that the electrostatic force on the tip is greater for the N-face GaN when no bias is applied and greater for the Ga-face GaN when the bias is greater than 1.5 V.

Taking into account the sign of the measured surface charge, the measured surface potentials and the dc bias that brought equivalence of the electrostatic force on the tip from the Ga- and N-face, the net surface charge density can be determined from the tip-sample capacitance, C_t, and the capacitance derivative, $\partial C_t/\partial z$, using the method of image charge approach.[12,13] Assuming the manufacturer specified tip radius $R=50$ nm and our experimental tip-sample distance $z=70$ nm, we obtain 7×10^{-18} F and -1.6×10^{-11} F/m for C_t and $\partial C_t/\partial z$, respectively.[12,13] The model used does not include the capacitance contributions from the cantilever beam, nor does it take into account the actual geometrical shape of the tip. Ignoring these effects, the net surface charge density difference can be determined to be $|\sigma^{\mathrm{N}}|-|\sigma^{\mathrm{Ga}}|=3.6\pm0.4\times10^{-5}$ C/m^2 prior to the surface treatment and $|\sigma^{\mathrm{N}}|-|\sigma^{\mathrm{Ga}}|=6.2\pm0.8\times10^{-5}$ C/m^2 after the surface treatment, indicating that there has been a net increase in the surface charge density difference between faces. Since the surface potential for the Ga-face remained the same, we attribute the change in the difference in surface charge density to be due to the N-face only. The corresponding increase in surface charge density for the N-face is roughly $1.6\pm0.5\times10^{10}$ electrons/cm^2, which is a small fraction of the bound polarization charge (2.12×10^{13} cm^{-2}). This slight modification of the surface charge has a negligible effect on the degree of screening, which is essentially 99.9% in either case. Since a reduction in net (positive) surface charge is observed and the electrostatic force on the tip changes only for the N-face GaN, it is reasonable to conclude that the surface treatment added adsorbed charge to the N-face regions.

SKPM measurements before and after the surface treatment revealed no change in the surface potential of the Ga-face regions, and a reduction in the surface potential of 0.3 ± 0.1 V for the N-face regions. Consider the surface potential difference $\Delta V_s=V_{\mathrm{N}}-V_{\mathrm{Ga}}=(1/e)(\Delta\phi_{\mathrm{Ga}}+\Delta\chi_{\mathrm{Ga}}-\Delta\phi_{\mathrm{N}}-\Delta\chi_{\mathrm{N}})$ between N- and Ga-face GaN with equal electron affinities and bulk Fermi level positions. If we attribute $\Delta\chi$ to a surface dipole between bound polarization charge and adsorbed screening charge, we expect that $\Delta\chi_{\mathrm{N}}$ acts to increase the electron affinity, while $\Delta\chi_{\mathrm{Ga}}$ acts to decrease the electron affinity. Since the surface treatment added charge to the N-face surface but not to the Ga-face surface, $\Delta\chi_{\mathrm{N-post}}>\Delta\chi_{\mathrm{N-pre}}>0$ while $\Delta\chi_{\mathrm{Ga}}$ remains unchanged. Therefore, $\Delta V_{\mathrm{pre}}-\Delta V_{\mathrm{post}}=(1/e)(\Delta\chi_{\mathrm{N-post}}+\Delta\phi_{\mathrm{N-post}}-\Delta\chi_{\mathrm{N-pre}}-\Delta\phi_{\mathrm{N-pre}})=0.3\pm0.1$ V. If we attribute this entirely to a surface dipole that changes the electron affinity of the surface, the 0.3 eV value would correspond to a charge density of $\sim1.6\times10^{13}$ cm^{-2} assuming a 1 nm dipole with a dielectric constant, $\varepsilon=10$ (corresponding to gallium oxide). This dipole charge is similar to the bound polarization charge but three orders of magnitude larger than the adsorbed charge that we observe. Therefore, it appears that in addition to a surface dipole, the HCl process must modify the band bending at the N-face surface.[14]

Considering all of the results here, the band bending at the as-received N-face surface is initially flat or slightly upward and increases as a result of the HCl clean. The deduced net charge is not large enough to account for the observed change in surface potential. Therefore, surface states or defects must be present near the surface to receive the excess negative charge to allow the upward band bending. Since these measurements were performed in air as opposed to a vacuum environment, it is difficult to establish the relative contribution from band bending and surface dipole.

In summary, EFM was used to determine the sign of the net surface charge, and to qualitatively determine the effect of an HCl surface treatment, while SKPM was used quantitatively to measure the contact potential difference before and after the surface treatment. The combination of EFM and SKPM allowed the difference in surface charge densities to be calculated. Unlike ferroelectric oxide surfaces, which have been found to be primarily screened by adsorbed species,[15] GaN is primarily screened by internal charge ($N_d=4.1\times10^{17}$ cm^{-3}). It has been found that the Ga-face surface was unaffected by the HCl surface treatment, while the surface potential of the N-face GaN was reduced in the process.

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