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Investigation of polarization-induced electric field in ultrathin InAlN layers on GaN by X-ray photoelectron spectroscopy

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The polarization-induced electric field in ultrathin $In_xAl_{1-x}N$ ($0.17 \leq x \leq 0.30$) layers on GaN was investigated by using X-ray photoelectron spectroscopy (XPS). The core-level energy position, E_{CL}, and the full width at half maximum (FWHM) of the Al2p, In4d, and In3d spectra from 2.5-nm-thick InAlN layers increased with the increase in the photoelectron exit angle (elevation angle). These increases were well reproduced with numerical calculations assuming polarization-induced internal fields combined with surface Fermi level pinning. The magnitudes of the internal field decreased as the In molar fraction increased. The Ga3d spectra from the host GaN layers markedly shifted by 530 meV depending on the molar fraction of InAlN layers, which was independent of the exit angle. This indicated that the Fermi level was unpinned at the interfaces, or GaN surfaces, and shifted due to the potential drop in the InAlN layers.

1 Introduction Lattice-matched $In_{0.17}Al_{0.83}N$/GaN heterostructures can provide a two-dimensional electron gas (2DEG) with high density exceeding 2×10^{13} cm^{-2} without doping [1–4], due to the difference in spontaneous polarization at the interface [5]. Piezoelectric polarization is added to spontaneous polarization in pseudomorphic $In_xAl_{1-x}N$ layers with a lattice mismatch to GaN. It has been reported that piezoelectric polarization increases as the In molar fraction increases to compensate the spontaneous polarization around x = 0.32 [5]. It is useful to experimentally confirm this feature for the device design. For this purpose, the polarization-induced electric field in ultrathin InAlN layers on GaN has to be investigated. One of the most useful methods of investigating ultrathin layers is X-ray photoelectron spectroscopy (XPS) because its probing depth is several nano-meters. We investigated the polarization-induced electric field in ultrathin lattice-matched and pseudomorphic $In_xAl_{1-x}N$ ($0.17 \leq x \leq 0.30$) layers on GaN by using XPS in this work.

2 Experimental The sample structure used to investigate InAlN ultrathin layers with XPS is schematically shown in Fig. 1. All the samples were grown by metal-organic vapor phase epitaxy (MOVPE) without doping. $In_{0.17}Al_{0.83}N$(2.5 nm)/ GaN, $In_{0.25}Al_{0.75}N$(2.5 nm)/ GaN, and $In_{0.30}Al_{0.70}N$(2.5 nm)/ GaN heterostructures were prepared. Here, we have defined the photoelectron exit angle, θ, as the elevation angle indicated in Fig. 1. Trimethylgallium was used as a precursor for gallium, trimethylaluminum for aluminum, trimethylindium for indium, and ammonia for nitrogen. The GaN, $In_{0.17}Al_{0.83}N$, $In_{0.25}Al_{0.75}N$, and $In_{0.30}Al_{0.70}N$ layers were respectively grown at 1000, 820, 780, and 730 °C. The details on the growth conditions are described in Ref. [6]. Successful control of the molar fraction in MOVPE growth was confirmed for the thick InAlN layers by using X-ray diffraction. XPS was done by using a monochromated Al-$K\alpha$ X-ray source (1486.6 eV). The binding energy was calibrated by adjusting the peak positions of the C1s core levels to 285.0 eV for each sample surface. The morphology of the surfaces was investigated by atomic force microscopy (AFM). The surfaces of all the heterostructure samples with the ultrathin InAlN layers were smooth (RMS roughness < 0.25 nm) which indicated that the thin InAlN layer had uniform thickness as

a result of layer-by-layer growth. The XPS data also indicated this state. Oxide layers at the sample surfaces were removed by pretreating them with hydro-fluoric (HF) acid.

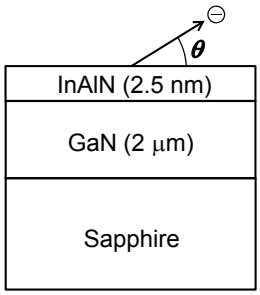

Figure 1 Structure of sample for XPS investigations.

3 Results and discussion Figure 2 shows anomalous behaviors observed for XPS Al2p spectra from $In_{0.17}Al_{0.83}N$ layers. The spectrum without band bending that was used for the numerical calculations (to be described later) is also plotted by the dotted line for comparison. For θ =15°, a slight amount of the oxide component was found by curve fitting and excluded. As can be seen, the core-level peak position, E_{CL}, and FWHM increased with an increase of θ. A similar situation was also observed for In3d and In4d spectra, while such θ–dependent increases were not observed for the Ga3d spectrum.

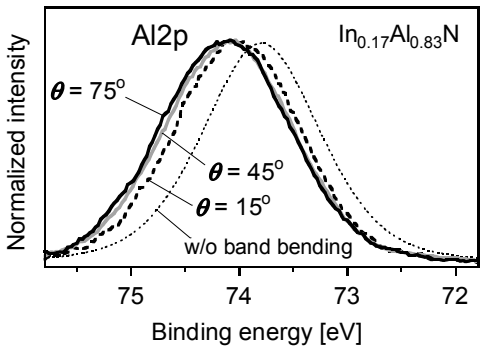

Figure 2 Observed Al2p spectra for $In_{0.17}Al_{0.83}N$(2.5 nm)/ GaN sample.

The observed increases in E_{CL} and FWHM indicated sharp band bending in the $In_{0.17}Al_{0.83}N$ layer. If there is band bending in the InAlN layer, core levels should also be bent along the band. The observed core-level spectrum, which is the integration of photoelectron responses from all depth points, then results in apparent increases in E_{CL} and FWHM. Numerical calculations, assuming polarization-induced internal fields combined with surface Fermi level pinning, were carried out to quantitatively interpret the observed increases. The theoretical basis for this is as follows. A core-level spectrum as a function of the binding energy, E, from a layer of thickness d can be given by [6]

$$I(E) = \int_0^d I_0(E,z)\exp\left(-\frac{z}{\lambda}\right)dz\,, \qquad (1)$$

where z, λ, and $I_0(E, z)$ indicate the depth from the surface, the escape depth of the photoelectrons, and the spectrum generated at each depth point. Here, the $I_0(E, z)$ for one spin orbital is represented by the pseudo-Voigt function. When surface band bending is not negligible on the scale of λ, the z-dependence of the core-level energy should be taken into account in $I_0(E, z)$. The λ can be changed by changing θ according to

$$\lambda = \lambda_0 \sin\theta\,, \qquad (2)$$

where λ_0 indicates the inelastic mean free path (IMFP) that was calculated using the equation in [7].

The actual values of the observed increases in E_{CL} and FWHM according to θ are plotted for Al2p, In4d, and In3d spectra from all the InAlN layers in Figs. 3(a)–(c) for comparison with the calculated results. The internal field assumed here was 3.2 MV/cm for $In_{0.17}Al_{0.83}N$, 2.4 MV/cm for $In_{0.25}Al_{0.75}N$, and 1.2 MV/cm for $In_{0.30}Al_{0.70}N$. The observed trend was in good agreement with the previously reported theoretical estimates [5]. The observed anomalous behaviors were well reproduced by the calculations for three spectra of all the samples.

More evidence of the internal field in InAlN layers was found in the E_{CL} of Ga3d spectra, E_{Ga3d}. The results are plotted by the closed circles in Fig. 4. The E_{Ga3d}, which was independent of θ, decreased as the In molar fraction increased. A difference in E_{Ga3d} as large as 530 meV was observed between $In_{0.17}Al_{0.83}N$/GaN and $In_{0.30}Al_{0.70}N$/GaN. The E_F positions from the valence band maximum, E_V, at the surface of the host GaN layers were evaluated by using the $(E_{Ga3d} - E_V)$ value of 17.1 eV, which was measured separately [8]. The absence of θ-dependence in E_{Ga3d} values meant that band bending in GaN was negligible within the range of the probing depth. The Fermi-level stabilization energy, E_{FS}, is also indicated by the broken line in Fig. 4. The E_F stabilizes at E_{FS} with high damage density [9]. The observed E_F positions further deviated from E_{FS} as the internal field in InAlN increased, which indicated that E_F was unpinned at the successfully formed heterointerfaces and shifted due to the potential drop in the InAlN layers.

4 Conclusion The polarization-induced electric field in 2.5-nm-thick $In_xAl_{1-x}N$ ($0.17 \le x \le 0.30$) layers on GaN was investigated by using XPS. The E_{CL} and FWHM of the core-level spectra from InAlN layers increased as θ increased. The observed increases were well reproduced by numerical calculations assuming the internal field due to the spontaneous and piezoelectric polarizations combined

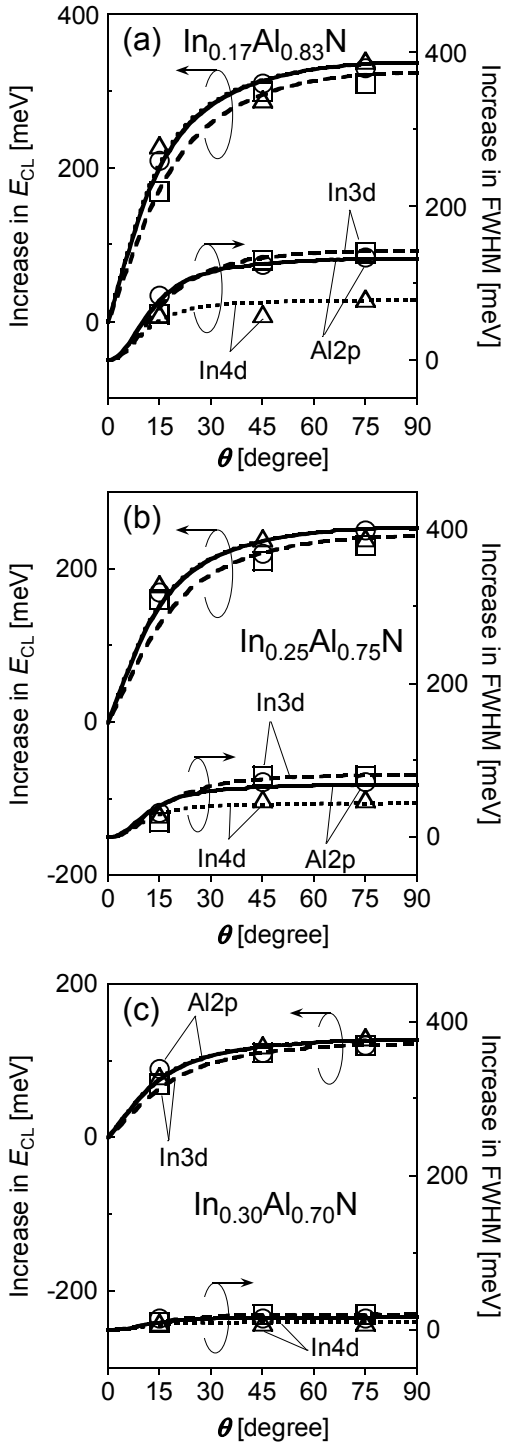

Figure 3 Observed apparent increases in E_{CL} and FWHM of Al2p, In3d, and In4d spectra. (a) $In_{0.17}Al_{0.83}N$(2.5 nm)/GaN, (b) $In_{0.25}Al_{0.75}N$(2.5 nm)/GaN, and (c) $In_{0.30}Al_{0.70}N$(2.5 nm)/GaN. Open circles, rectangles, and triangles indicate observed increases for Al2p, In3d, and In4d, respectively. Solid line, broken line, and dotted line plot calculated results for Al2p, In3d, and In4d, respectively.

with surface Fermi-level pinning. A difference in E_{Ga3d} as large as 530 meV was observed between $In_{0.17}Al_{0.83}N$/ GaN and $In_{0.30}Al_{0.70}N$/GaN, which was independent of θ. This indicated that E_F was unpinned at the interfaces and shifted due to the potential drop in the InAlN layers.

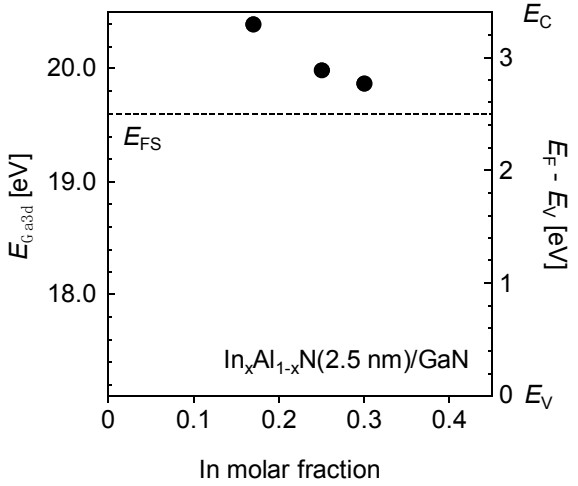

Figure 4 Observed Ga3d peak position and evaluated E_F position at the surface of GaN beneath 2.5-nm-thick InAlN layers. Broken line indicates E_{FS} level for GaN.

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