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COMMUNICATIONS

Polarity determination of a GaN thin film on sapphire (0001) with x-ray standing waves

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The x-ray standing wave technique was used to determine the polarity of a 1 μm thick GaN film grown by molecular beam epitaxy on an α-$Al_2O_3(0001)$ single crystal. The standing wave was generated by x-ray diffraction from the GaN film. The Ga K_α fluorescence yield was recorded as a function of incidence angle within the range of the GaN(0002) reflection. Analysis of the data reveals that the film has grown with N polarity, i.e., the nitrogen atoms occupy the top half of the wurtzite (0001) bilayers. © *1998 American Institute of Physics.* [S0021-8979(98)08115-8]

Noncentrosymmetric compound crystals exhibit two different sequences of the atomic layering in the two opposing directions parallel to certain crystallographic axes, and consequently crystallographic polarity along these axes can be observed. For binary $A-B$ compounds with wurtzite structure, the sequence of the atomic layers of the constituents A and B is reversed along the $[0001]$ and $[000\bar{1}]$ directions. The corresponding (0001) and $(000\bar{1})$ faces are terminated by atoms A or atoms B, respectively, and the polarity strongly influences the chemical and physical properties of the exposed polar surface. In the case of heteroepitaxial growth of thin films of a noncentrosymmetric compound, the polarity of the epilayer cannot be predicted in a straightforward way, and an experimental study is needed. This is the case for wide band-gap semiconductor GaN films which are attracting interest in their usage as short-wavelength light emitting diodes and lasers. Both types of polarity were reported to be found by ion channeling and convergent beam electron diffraction in GaN(0001) layers grown by metalorganic chemical vapor deposition (MOCVD) on sapphire (0001) if the layers exhibited rough morphology, while for smooth films Ga termination was exclusively concluded from the experimental results.[1] This result was supported very recently by a photoelectron diffraction study of MOCVD grown films.[2] In this communication we describe a novel experimental approach to the determination of the polarity of a GaN epitaxial film employing the x-ray standing wave (XSW) technique. The 1 μm thick GaN film was grown by molecular beam epitaxy (MBE) on an α-$Al_2O_3(0001)$ single crystal. In contrast to the above mentioned studies of MOCVD grown samples, we find that our high-quality MBE grown film has N polarity.

Apart from different chemical etching techniques, several methods have been developed and used to determine the face polarity of noncentrosymmetric crystals, such as x-ray diffraction,[3] Auger electron spectroscopy,[4] electron diffraction,[5] and ion channeling.[6] The uniqueness of the XSW technique[7] lies in the combination of the structural sensitivity of x-ray diffraction and the chemical elemental sensitivity inherent to x-ray spectroscopy. The method is typically based on generating an XSW field by x-ray Bragg diffraction and monitoring the x-ray fluorescence yield excited by this field as a function of glancing angle θ as the crystal is tuned through the narrow region of Bragg reflection either by varying θ or the x-ray energy E_γ. During the past 2 decades, the XSW method has matured to a powerful tool for investigating the structure of adsorbates, interfaces, and thin films.[8] Determining the polarity of noncentrosymmetric crystals with the XSW technique is straightforward and has been demonstrated for single crystals such as GaP,[9,10] CdS,[11] GaAs,[11,12] and CdTe.[13]

The basis for using the XSW method to determine the polarity is illustrated by Fig. 1 for the case of GaN(0001). Approaching the angular region of Bragg reflection from the low angle side, the XSW field is formed with its maxima, i.e., the *antinodal* planes, positioned between GaN bilayers. By tuning the crystal through the diffraction region, the maxima of the XSW move *inward* in the $-\mathbf{H}$ direction by one-half of the XSW period ($d_{\mathrm{xsw}}=d_{0002}$); the antinodes of the XSW cross the upper half of the bilayer at the high angle side, and are finally positioned between the Ga and N atomic

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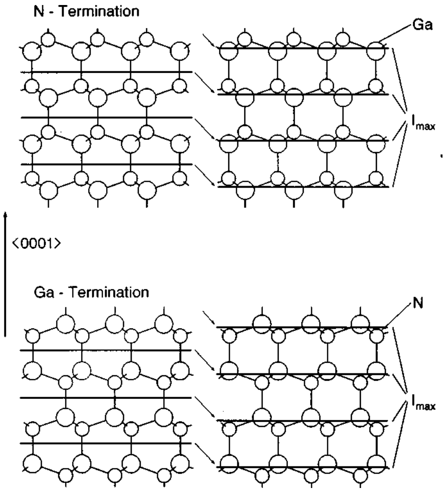

FIG. 1. The application of the XSW method for polarity determination of (0001) oriented GaN sketched in side view; shown is the movement of the maxima of the XSW field, proceeding from the low angle side (left-hand side) to the high angle side of the Bragg reflection range (right-hand side). The movement of the maxima of the wave field intensity ($I_{\max}$), i.e., the antinodes, is shown for N termination (top) and Ga termination (bottom).

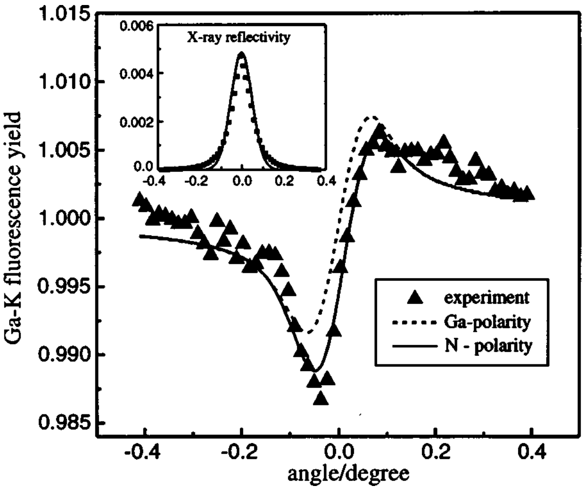

FIG. 2. Experimental Ga *K* fluorescence yield curve (filled triangles) from a 1 μm GaN film on sapphire(0001). (Solid line) the best fit corresponding to N polarity of the film and a static Debye–Waller factor of $e^{-W}=0.36$. The calculated best fit for the Ga polarity is also shown for comparison (the dashed line). The experimental reflection curve is shown in the inset (squares) together with a fit (solid line) by convoluting the theoretical curve with a Gauss function with $\sigma=0.05°$.

planes of the bilayer, i.e., at the diffraction plane position,[14–16] as shown on the right-hand side of Fig. 1. Outside the diffraction region, the XSW vanishes, and the x-ray intensity is constant for all positions within the GaN unit cell. As we can see in Fig. 1, for the N-terminated case, as θ increases through the reflection, each XSW antinode moves away from an upper Ga plane as it approaches a lower Ga plane, which it never reaches. In the case of Ga termination, each antinode does pass through a Ga plane as θ is increased through the (0002) reflection. As a consequence, the Ga fluorescence yield will reach its maximum value earlier than in the N-terminated case and the maximum will also reach a higher value.

Perfect single crystals exhibit intrinsically very narrow reflectivity curves, as described by the dynamical theory of x-ray diffraction.[14] With decreasing perfection of the crystalline lattice the reflectivity curves broaden and eventually it is not possible to make reliable XSW measurements, which is unfortunately the case for many new and interesting materials. However, by decreasing the crystal thickness below the extinction length,[14] the width of the reflection curve becomes approximately inversely proportional to the thickness. This also leads to a reduction of the reflectivity R, but the intensity of the XSW field is proportional to $\sqrt{R}$.[17] Even for R as low as 10^{-4}, the modulation of the fluorescence yield, produced by the standing wave, is measurable if an intense synchrotron radiation source is employed. The XSW analysis, employing Bragg reflection from thin films, has been used successfully in the past.[18–20] In this communication we generate an XSW field within a 1 μm thick GaN film.

The epitaxial GaN film was grown on an $Al_2O_3(0001)$ substrate without a buffer layer by plasma induced molecular beam epitaxy (PIMBE) using a conventional gallium effusion cell and a rf plasma atomic radical source for nitrogen. The inductively coupled plasma power was 400 W and the pressure in the MBE chamber during growth was 4×10^{-5} mbar. The gallium flux and the substrate temperature during growth, including the initial stage, were fixed at 8×10^{14} cm^{-2}s^{-1} and 810 °C, resulting in a calibrated growth rate of 0.6 μm/h.[21]

The XSW experiment was performed at the X15A beamline of the National Synchrotron Light Source, BNL. An x-ray energy of 10.5 keV was selected by the double crystal Si(111) monochromator, thus slightly above the Ga *K* edge (10.372 keV). The XSW measurement was performed employing the (0002) reflection from the wurtzite structure GaN thin film. An energy-dispersive Si(Li) detector was used to record the Ga *K* fluorescence photons from the epilayer. The results of the measurement are shown in Fig. 2.

To fit the experimental reflectivity curve, the calculated reflection curve was convoluted with a Gaussian function to account for broadening of the curve due to defects in the film. The best fit gives $\sigma=0.05°$ (inset in Fig. 2). This value was fixed and used for the convolution of the calculated fluorescence yield. For the calculations of the XSW curve, we used the algorithm developed in Ref. 22 for multilayer crystalline systems. The experimental fluorescence yield curve was fitted for both cases—N and Ga polarity of the film—using only the static Debye–Waller factor e^{-W} as a fitting parameter. With the static Debye–Waller factor we take crystalline imperfections of the film structure into account, with $e^{-W}=1.0$ for a perfect film and $e^{-W}=0$ for a completely disordered, amorphous layer. Already by visual inspection of the fitted curves for N and Ga polarity in Fig. 2, it is obvious that our film exhibits N polarity. Figure 3 is a measure of the reliability of our polarity determination; it shows the χ^2 values from the least-squares fit for both polarities as a function of e^{-W}. One can see that both χ^2 curves display the minimum at $e^{-W}=0.36$, with the best χ^2 value for the N polarity less than one fourth of that for the Ga polarity. Assuming a mixture of both types of polarity, the fit

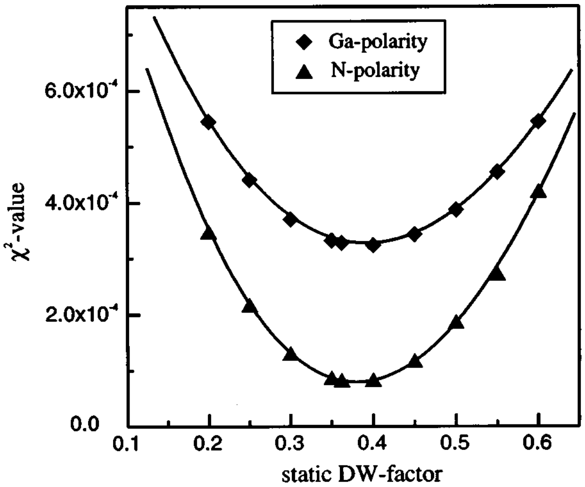

FIG. 3. χ^2 values as a function of the fitting parameter e^{-w} for Ga polarity (diamonds) and N polarity (triangles) of the film. The χ^2 value for Ga polarity is almost five times worse than for N polarity.

becomes worse. N polarity exclusively yields the lowest χ^2 value. The XSW curve corresponding to the best fit is shown in Fig. 2 (the curve for the Ga polarity is also shown for comparison as a dashed line).

In conclusion, we successfully applied the XSW technique to determine the polarity of a noncentrosymmetric epitaxial film, using the 1 μm thin film as a standing wave generator. Our results are in contrast with recent reports[1,2] where Ga polarity was found for flat GaN films grown on sapphire single crystals by MOCVD. In these studies N polarity was found as a minority phase under slightly varied growth procedures, whereas for our MBE grown film we find N polarity predominantly; the margins of error may allow for a 10% fraction with Ga polarity.[23] Since the sharpness of the Bragg reflection of our MBE-grown film testifies to its high quality, our results prove that excellent GaN films of both polarities can be grown depending on the choice of the growth procedure.

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