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Optical properties of GaN grown on ZnO by reactive molecular beam epitaxy

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High quality wurtzite GaN epilayers have been grown on ZnO(0001) substrates by reactive molecular beam epitaxy. Photoluminescence and reflectivity measurements point to high quality presumably due to the near match of both the crystal lattice parameter and the stacking order between GaN and ZnO. In addition, the good films lack the characteristic yellow photoluminescence band. Any misorientation of the GaN epilayer planes with respect to the ZnO substrate is not detectable with polarized reflectivity. The x-ray double crystal diffraction measurements indicate this misorientation is much smaller than those for GaN epilayers on SiC and Al₂O₃. © 1997 American Institute of Physics. [S0003-6951(97)01304-1]

GaN is a wide gap semiconductor with great promise for optoelectronic and high-temperature/high-power device applications.¹⁻⁴ The challenge of growing high quality and defect-free GaN and its alloys, In_{1-x}Ga_xN and Al_{1-x}Ga_xN, is still formidable due to the lack of lattice matched substrates. Most of the previous work has been devoted to the growth of GaN on Al₂O₃ by metal organometallic vapor pressure epitaxy (MOVPE)^{2,3} or molecular beam epitaxy (MBE).⁵ Despite the demonstration of light emitting diode² and pulsed laser devices³ grown on Al₂O₃, the density of defects is probably still too high² to achieve the desired violet and blue lasers that operate continuously at room temperature with the required longevity, as well as high-power electronic devices.⁴

Successful MBE and MOVPE growth of wurtzitic GaN has been demonstrated on various substrates such as 6H-SiC,⁶ MgAl₂O₄,⁷ Si(111),⁸ GaAs,⁹ and ZnO(0001).^{10,11} Sverdlov *et al.*⁶ discussed the possible causes of the dense network of threading defects in epitaxial hexagonal GaN films grown on nonisomorphic substrates and have suggested that the stacking mismatch between the substrate and the epitaxial layer is responsible for many of these defects. Transmission electron microscopy (TEM) experiments^{6,11} have revealed the presence of planar defects in GaN epitaxial layers grown on Al₂O₃ and SiC. Two types of defects have been considered: double positioning boundaries, also called stacking mismatch boundaries (SMB), and inversion domain

boundaries (IDB). In the cases of SMB there is an equal probability of nucleating two different face-centered-cubic (fcc) stacking sequences (*ABC* and *ACB*). For example, the stacking sequence in 6H-SiC is *ABCACB*, so that three stacking sequences would be possible for wurtzite GaN: substrate *ABC* leading to *BCBC* GaN; substrate *BCA* leading to *CACA* GaN; and substrate *CBA* leading to *BABA* GaN.

ZnO is being considered as a suitable substrate for GaN growth because of its stacking order match and close lattice match. The lattice mismatch between GaN and ZnO is $\epsilon = 0.017$, which leads to a range of critical thickness between 80 and 120 Å, estimated using different elastic models.¹² This implies that coherently strained layers of GaN could be pseudomorphically grown with thickness up to about 100 Å on ZnO substrates. Since the steps on (0001) 2H ZnO would be bilayer ones, SMB-type defects would not be expected to occur. Matsuoka *et al.*¹⁰ have used Zn-face ZnO substrates to grow the lattice matched alloy In_{0.22}Ga_{0.78}N. More recently, ZnO has been used as a compliant buffer layer^{13,14} in the growth of high quality GaN. Nevertheless, the ZnO buffer layers grown on substrates with which they do not share a common stacking order suffer from stacking faults that would cause the GaN films grown on them to still have high density of SMBs. It should, therefore, not come as a surprise that the quality of epitaxial GaN grown on compliant buffer layers is still inferior to the quality of the buffer layer used.¹⁴ In this letter, we present results of GaN grown, by reactive MBE,⁵ on high quality bulk ZnO(0001) substrates.

Figure 1 shows the 4.2 K photoluminescence (PL) and the reflectivity data obtained on a ZnO(0001) substrate. The

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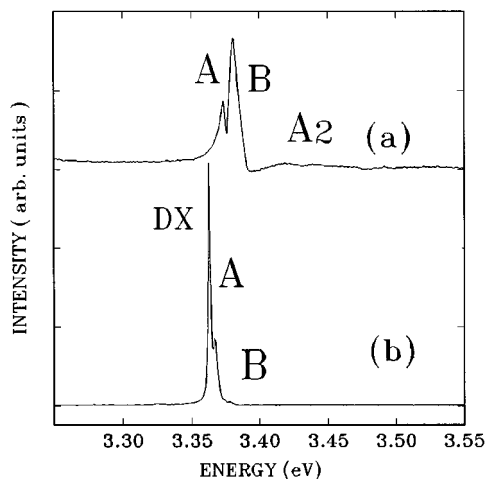


FIG. 1. (a) Reflectivity and (b) photoluminescence spectra of ZnO(0001) substrate at 4.2 K and with light configuration $k\parallel c$ and E perpendicular to c . Labels A, B, A_2 , and D^0X are given as identifications of free and bound excitons, respectively.

positions of the free and bound excitons are given as follows: A-exciton (3375 meV), B-exciton (3386 meV), and D^0X (3364 meV). The PL peaks have a full width at half-maximum (FWHM) of 3 meV, which indicate a reasonably high quality ZnO substrate. The observation of the A and B free excitons using different polarized light confirms the orientation of the substrate. However, the substrates suffer from rather poor surface preparation, limiting the quality of the GaN films. We note that the first excited A_2 exciton is also observed in the reflectivity spectrum of the ZnO substrate itself, and that the energy position of A_2 exciton is consistent with the high exciton binding energy of 67 meV in ZnO. The observation of the excited free exciton indicates that the substrate has low carrier concentration and very good crystalline quality.

Figure 2 shows PL and reflectivity spectra of GaN epilayers grown by reactive ammonia MBE on ZnO, at 780 °C. The reflectivity spectrum shows pronounced features at

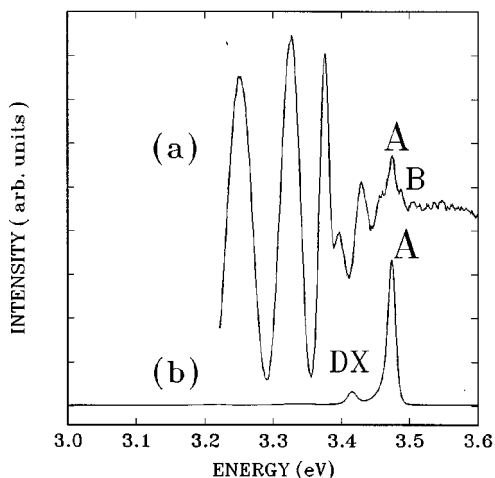


FIG. 2. (a) Reflectivity and (b) photoluminescence spectra of GaN/ZnO(0001) at 4.2 K and with light configuration $k\parallel c$ and E perpendicular to c . Labels A, B, and D^0X are given as identifications of free and bound excitons, respectively.

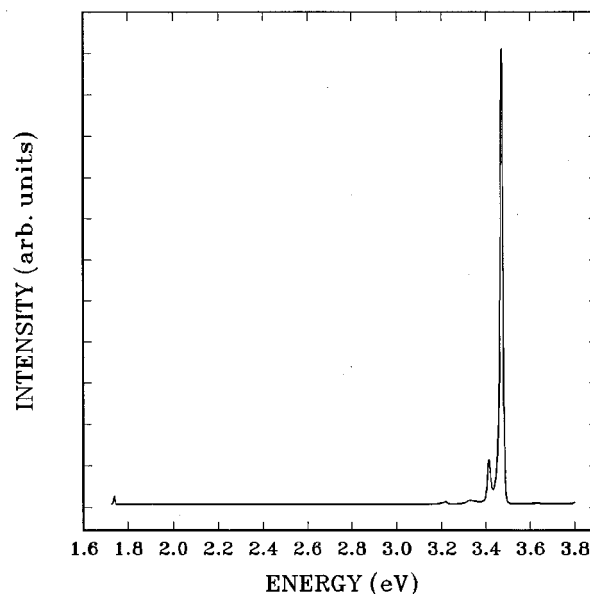


FIG. 3. Photoluminescence spectrum of GaN/ZnO at 4.2 K. The yellow band is not observed at either room temperature or 4.2 K, even with a high detection sensitivity (wide open monochromator slits, 600 μm).

higher energy corresponding to the A (3480 meV) and B (3493 meV) free excitons. The oscillations observed in the reflectivity spectrum at lower energies are due to interference fringes between the reflections from the surface of GaN and the GaN/ZnO interface. The fringe period gives an estimate of the GaN thickness at about 1.5 μm , which is consistent with the growth rate of 0.8 $\mu\text{m}/\text{h}$. It is worth noting that the discontinuity in the oscillation fringes occurring in the range between 3.38 and 3.45 eV is due to the spectral dispersion of the refractive index of ZnO close to the energy gap. The PL spectra show a peak at the same energy position as the A exciton in the reflectivity spectra, which allowed us to attribute this peak to the free A exciton. Despite the rather poor substrate surface preparation, the PL and reflectance widths of the excitonic transitions (FWHM ~ 8 meV) are approximately in the same range of the best results obtained on sapphire substrates by MBE. It is very important to note the lack of any yellow band luminescence in the room and low-temperature PL spectra of the samples grown on ZnO (0001) substrates. As shown in Fig. 3, the near band-edge transitions, i.e., free and shallow bound excitons, are the only transitions observed in these samples. Even at a high detection sensitivity (wide open monochromator slit width of 600 μm), the yellow band was completely absent.

Recent first-principles calculations of domain wall energies for both IDB and SMB in GaN (Ref. 15) indicate that the IDB does not induce electronic states in the band gap, but that SMBs give rise to occupied N-derived interface states in the band gap. Therefore, one source of the commonly observed yellow band, may be the SMBs.¹⁵ Other likely sources may have to do with electron-hole recombination processes involving impurities or native point defects such as deep acceptor levels (Ref. 16) and Ga vacancies.¹⁷ Consequently, an argument can be made that the absence of any yellow PL signal in our GaN samples grown on ZnO, as shown in Fig. 3, is an indication of a reduced density of

defects. TEM results on our GaN grown on ZnO indicate much improved quality near the surface over those on sapphire, and defects at the epilayer–substrate interface caused by poor ZnO surface preparation. Atomic force microscopy results indicate that GaN grown on ZnO presents a surface roughness comparable to GaN grown on Al₂O₃.

The strain induced both by lattice mismatch and the difference between the expansion coefficients of the layers and the substrate induces a shift and an alteration of the excitonic transitions in GaN. Gil *et al.*¹⁸ have presented a theoretical model using the Pikus and Bir Hamiltonian, taking into account the relaxation of the thermal and lattice mismatch between GaN and the substrate, in order to fit the energy shift of both the conduction and valence bands. It is noted that GaN layers grown on sapphire undergo compressive strain, while GaN layers grown on SiC are under tensile strain. An accurate estimate of the thermal strain (ε_{th}) due to the difference between the thermal expansion coefficients of GaN and the particular substrate is given by

$$\varepsilon_{th} = [\Delta a_l(T) - \Delta a_s(T)] / \Delta a_s(T), \quad (1)$$

where $\Delta a_l(T)$ and $\Delta a_s(T)$ represent the variation of the lattice parameter between the growth temperature and room temperature for the GaN layer and the substrate, respectively. Our estimate of ε_{th} , using the temperature dependence of the thermal expansion coefficient¹⁹ gives a positive value for ε_{th} for both sapphire and ZnO substrates and a negative value for SiC substrates. This is consistent with the energy position of the free exciton observed in both reflectivity and photoluminescence spectra.¹⁸ Note that the compressive thermal strain induced by a ZnO substrate is smaller than that induced by sapphire substrates. This implies that the cracking of GaN layers observed in growth on Al₂O₃ and SiC substrates could be avoided where ZnO is used as a substrate. In accordance with the optical data, x-ray double crystal diffraction results obtained in samples grown both on ZnO and sapphire indicate that the value of the lattice parameter perpendicular to the interface of GaN grown on ZnO is closer to that of bulk GaN.

The intensity of reflectance peaks corresponding to the free exciton transitions is a function of their oscillator strengths.²⁰ Using linearly polarized reflectance measurements on GaN grown on sapphire, we have observed a variation of the exciton oscillator strength, which indicates that such GaN layers are tilted with respect to the substrate axis. Our results for GaN samples grown on ZnO indicate an absence of any tilting effect. Double crystal x-ray diffraction measurements performed by rotating the samples 180° show a smaller variation in the peak position for growth on ZnO than for growth on SiC or Al₂O₃.

In conclusion, photoluminescence and reflectance data show that high quality GaN epilayers can be grown on ZnO(0001) substrates using reactive ammonia MBE. The absence of the yellow photoluminescence band indicates a reduced defect density in GaN grown on ZnO substrates. X-ray measurements as well as polarized optical data confirm that the GaN epilayers are well oriented with respect to the substrate axis. The lower strain offered by ZnO substrates may eliminate cracks occurring for growth on sapphire and SiC substrates. Further investigations on the effect of surface polarity and different buffer layers are under way.

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