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Polarization-dependent spectroscopic study of *M*-plane GaN on γ-$LiAlO_2$

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We investigate the polarization dependence of the absorption, reflectance, and photoreflectance spectra of a compressively strained, *M*-plane, wurtzite GaN($1\bar{1}00$) film grown by molecular-beam epitaxy on a γ-$LiAlO_2$(100) substrate. The measurements are done with the electric-field vector (**E**) of the probe light being parallel (∥) and perpendicular (⊥) to the **c** axis of GaN, which lies in the growth plane. We observe a significant increase in the effective optical band gap of the *M*-plane GaN film for **E**∥**c** compared to its value for **E**⊥**c**. This result is explained by including the effect of the *M*-plane biaxial compressive strain on the electronic band structure of GaN. We also determine the extraordinary refractive index of GaN at energies below its band gap from the reflectance measurements. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1434306]

The performance of GaN-based optoelectronic devices has improved significantly in recent years. Lately, however, it has been realized that due to the wurtzite (WZ) structure of GaN spontaneous and piezoelectric polarization related electrostatic fields arise when the growth direction of nitride heterostructures is [0001] oriented (*C*-plane film).[1] So far, devices have been made mostly on substrates that favor *C*-plane growth. The electrostatic fields tend to separate the electron and hole wave functions in a quantum well, reducing their overlap and resulting in lower device efficiency.[2] Recently, it has been demonstrated that such electrostatic fields can be avoided by growing [$1\bar{1}00$]-oriented (*M*-plane) GaN films.[3] In this case, the unique **c** axis of WZ GaN lies in the growth plane, leading to an anisotropy in the optical properties of the film for the light electric-field vector (**E**) polarized parallel (∥) and perpendicular (⊥) to the **c** axis. Additionally, if the film experiences strain, modification of the electronic band structure (EBS) is expected, resulting in further changes in the optical properties. Therefore, understanding of the polarization dependence of the optical properties of *M*-plane GaN films is of crucial importance for optoelectronic device applications.

In this letter, we investigate the optical polarization anisotropy of an *M*-plane GaN film under in-plane biaxial compressive strain. We provide an explanation for the observed polarization-dependent band-gap change. We also determine values of the extraordinary refractive index for GaN at energies below its band gap.

The GaN film used in this study was grown by rf plasma-assisted molecular-beam epitaxy on a γ-$LiAlO_2$(100) substrate.[4] The film thickness of 1.22 μm was accurately determined by scanning electron microscopy. The unintentional *n*-type background doping density was measured to be $5\times10^{17}\text{cm}^{-3}$ at 295 K. High-resolution triple-axis x-ray diffraction (XRD) and Raman spectroscopy were used to verify the single phase nature and *M*-plane orientation of the film.[4] The XRD measurement performed at 295 K reveals that the film is under biaxial compressive strain with an out-of-plane dilatation $\epsilon_{yy}=0.29\%$, which is expected to increase at 5 K by less than 10%. The width of the GaN($1\bar{1}00$) XRD-reflection peak is larger than theoretically expected, indicating a certain inhomogeneity of the strain distribution. The compressive strain observed is in agreement with Raman measurements, which show strongly blueshifted signals. The probe beam for photoreflectance (PR), reflectance (R), and transmission measurements was obtained by dispersing light from a Xe lamp using a 0.64 m monochromator and polarizing it with a Glan–Taylor prism. A He–Cd laser (3.815 eV) was used as the pump beam for PR.

Figure 1 shows the absorption spectra of the sample for **E**⊥**c** (**E**∥**x**) and **E**∥**c** (**E**∥**z**) at 295 and 5 K, with the light propagation direction being along **y**∥[$1\bar{1}00$], which is also the growth direction. The absorption spectra were obtained from fitting theoretical formulas[5] for transmission through a GaN film on a $LiAlO_2$ substrate to the measured transmission spectra. The parameters for $LiAlO_2$ were taken from Ref. 6. An example is shown in the inset of Fig. 1. We find

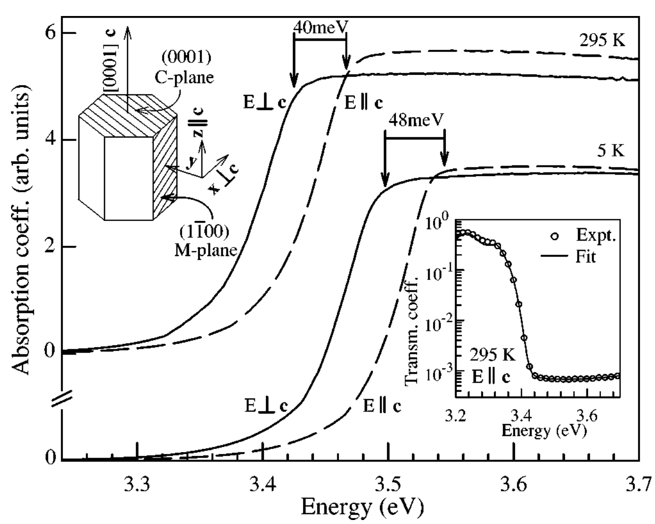

FIG. 1. Absorption spectra of the *M*-plane GaN film for **E**⊥**c** (solid lines) and **E**∥**c** (dashed lines). The arrows indicate the PR transition energies. The inset shows a typical transmission spectrum, from which the absorption coefficient was obtained. The wurtzite GaN unit cell and the choice of coordinates is also shown as a schematic in the inset.

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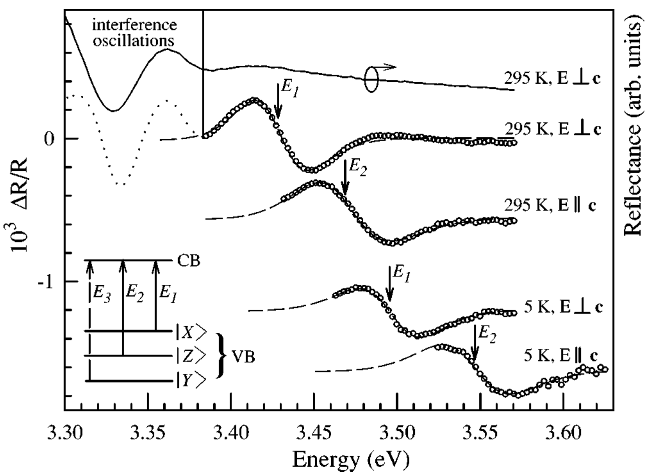

FIG. 2. Experimental PR spectra (circles) of the M-plane GaN film for $\mathbf{E}\perp\mathbf{c}$ and $\mathbf{E}\|\mathbf{c}$. The dashed lines are fits to Eq. (1) extrapolated to lower energies. Prior to the fits, the low energy part of the PR spectra was cut off at the energy, coinciding with the start of interference fringes in the R spectra. The R spectrum for $\mathbf{E}\perp\mathbf{c}$ at 295 K is shown as an example. The schematic inset shows the calculated band alignment in GaN for compressive strain within the M plane with ε_{xx}, $\varepsilon_{zz}\lesssim -0.2\%$.

that, when the polarization of the light is rotated by 90° from $\mathbf{E}\perp\mathbf{c}$ to $\mathbf{E}\|\mathbf{c}$ in the M plane, the spectrum is shifted toward higher energy, indicating an increase of the effective band gap.

Figure 2 displays PR spectra of the sample for both polarizations at 295 and 5 K. For each polarization, the spectra consist of a single resonance, whose energy depends on the polarization direction, just as in case of the absorption spectra. The nearly temperature-independent line shapes and the absence of sharp excitonic resonances in the absorption spectra are likely due to the dominance of inhomogeneous broadening related to the inhomogeneous strain distribution and the background doping density mentioned above. To obtain the transition energies (labeled E_1 and E_2), we fit the spectra using Aspnes' line shape function

$$\frac{\Delta R}{R} = \mathfrak{Re}\left[\frac{Ae^{i\Theta}}{(E-E_j+i\gamma)^m}\right], \tag{1}$$

with the exponent $m=3$ representing a Gaussian-broadened excitonic transition. The fitting parameters A, E_j, γ, and Θ denote the amplitude, transition energy, broadening parameter, and phase factor, respectively. The transition energies obtained are $E_1=3.498$ eV (3.428 eV) and $E_2=3.546$ eV (3.468 eV) at 5 K (295 K). The statistical error of the fits is ± 3 meV for all values. We conclude that the effective band gap of the M-plane film increases by 48 meV (40 meV) at 5 K (295 K) with an error of ± 6 meV, when the polarization of the light is rotated by 90° from $\mathbf{E}\perp\mathbf{c}$ to $\mathbf{E}\|\mathbf{c}$.

Unstrained wurtzite GaN has three closely spaced top valence bands (VBs) at the Brillouin-zone center (BZC): Γ_9 [heavy hole (HH)], upper Γ_7 [light hole (LH)], and lower Γ_7 [crystal field split-off hole (CH)]. Excitons involving electrons in the conduction band (CB) and holes in these VBs are referred to as A, B, and C excitons, respectively. The VB is constructed of p orbitals with wave functions mainly of type $|X\pm iY\rangle$ for the HH and LH bands and of type $|Z\rangle$ for the CH band.[7] The $\mathbf{c}$ axis defines the $\mathbf{z}$ direction. Under C-plane biaxial compressive strain with $\epsilon_{xx}=\epsilon_{yy}$, the near equivalence between the HH and LH wave functions remains unaffected. Therefore, in unstrained or compressively strained C-plane GaN, one expects to see two dominant transitions (A and B) for $\mathbf{E}\perp\mathbf{c}$ and one dominant transition (C) for $\mathbf{E}\|\mathbf{c}$. In effect, a polarization-dependent band-gap change, equal in magnitude to the difference between the lowest energy A and highest energy C transition, would occur. However, this has never been observed experimentally, because it is difficult to get pure $\mathbf{E}\|\mathbf{c}$ polarization with a C-plane GaN film.[8,9]

With an M-plane film, both pure $\mathbf{E}\perp\mathbf{c}$ and pure $\mathbf{E}\|\mathbf{c}$ polarizations are possible, but it would be incorrect to associate E_1 with an unresolved A-B and E_2 with the C exciton transition. This is because theoretical studies by Domen *et al.*[10] suggest that strain within the M plane modifies the EBS differently from strain within the C-plane case in that it removes the equivalence between the HH and LH bands. For a direct comparison with experiment, we have performed a $\mathbf{k}\cdot\mathbf{p}$ EBS calculation that includes the influence of strain.[11] The results show that for M-plane biaxial compressive strains $\epsilon_{xx},\epsilon_{zz}\lesssim -0.2\%$ the three VB states at the BZC have a predominantly $|X\rangle$-, $|Y\rangle$-, and $|Z\rangle$-like character. The $|X\rangle$-like VB is aligned closest in energy to the CB, then the $|Z\rangle$-like VB, and finally the $|Y\rangle$-like VB (cf. the inset of Fig. 2). We identify E_1, which has the lowest energy and is seen for $\mathbf{E}\perp\mathbf{c}$ ($\mathbf{E}\|\mathbf{x}$), with transitions to the $|X\rangle$-like VB. E_2, at higher energy and seen for $\mathbf{E}\|\mathbf{c}$ ($\mathbf{E}\|\mathbf{z}$), is identified with transitions to the $|Z\rangle$-like VB. By comparing the experimental E_1 and E_2 values at 5 K with the EBS calculations, we can estimate the in-plane strain ϵ_{xx} as well as ϵ_{zz} and therefore the dilatation ϵ_{yy}.[12] By varying only the deformation potential D_5 by $\sim 15\%$ from the theoretically predicted value by Ohtoshi *et al.*,[13] we were able to obtain the experimental value of $\epsilon_{yy}=0.29\%$ from $\epsilon_{xx}=-0.56\%$ and $\epsilon_{zz}=-0.31\%$. Although a polarization-dependent, effective band-gap change is possible in principle for unstrained GaN as discussed above, the polarization properties of the features seen here together with their energies can only be explained by including the effect of M-plane biaxial compressive strain on the EBS of GaN. Note also that for unstrained GaN or for compressively strained C-plane GaN the band-gap change would involve the lowest (A exciton) and highest (C exciton) energy transitions, while here it involves the lowest (E_1) and next higher (E_2) energy transition. In our case for PR, it is not possible to observe the highest energy (E_3) transition to the $|Y\rangle$-like VB (expected at 3.58 eV), since a sizeable $\mathbf{E}\|\mathbf{y}$ polarization is not achievable with an M-plane film. In a polarized photoluminescence study of M-plane GaN grown on $(1\bar{1}00)6H$-SiC, shifts in the emission peak positions were also reported,[14] but this was not attributed to a strain-induced EBS modification.

Figure 3(a) shows the normal-incidence R spectrum for the two polarizations. The shift between the interference oscillations seen in the two spectra is due to the difference between the value of the ordinary n_o (for $\mathbf{E}\perp\mathbf{c}$) and the extraordinary n_e (for $\mathbf{E}\|\mathbf{c}$) refractive index of GaN. Thus, the M-plane orientation of the film enables measurement of n_e, which is more intricate for C-plane samples.[15–18] An enlarged view of the band-gap region [inset of Fig. 3(a)] shows a polarization dependent shift in the damping of the interference oscillations, which is associated with the onset of ab-

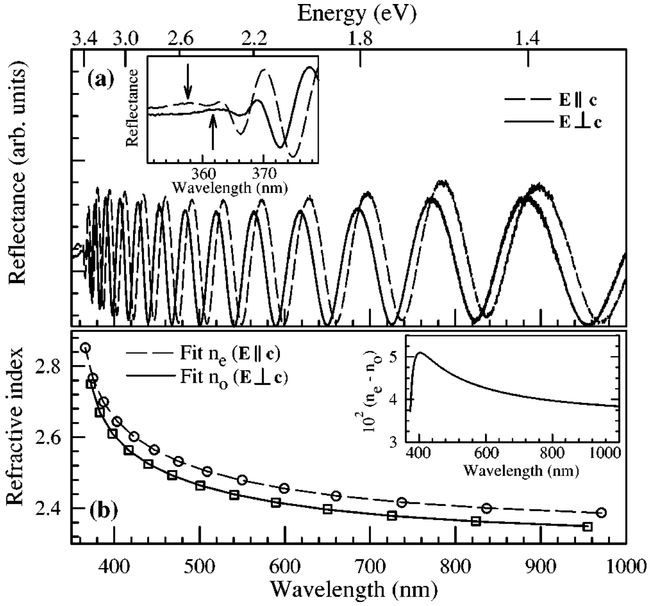

FIG. 3. (a) R spectra of the M-plane GaN film for **E**⊥ **c** and **E**∥**c** at 295 K. The inset shows an enlarged view around the band gap, and the arrows mark the PR transition wavelengths. (b) Measured refractive index n_0 (□) and n_e(○). The lines are fits using Eq. (2). The inset shows the dispersion of $n_e - n_0$.

sorption. This is in agreement with the polarization-dependent band-gap change discussed above. n_o and n_e are determined from the wavelength positions of the interference oscillation extrema. Sellmeir's relation of the form[5]

$$n^2(\lambda) = \varepsilon_\infty + \frac{a_1}{\lambda^2 - b_1^2} + \frac{a_2}{\lambda^2 - b_2^2} \tag{2}$$

with wavelength λ was fit to the dispersion measured [cf. Fig. 3(b)]. For n_e, the parameters ε_∞, a_1, b_1, a_2, and b_2 have values of 5.514, 0.1471 μm^2, 0.1482 μm, 0.018 88 μm^2, and 0.346 μm, respectively. For n_o, the above parameters were found to be 5.346, 0.1377 μm^2, 0.1573 μm, 0.014 92 μm^2, and 0.3524 μm, respectively. There is a systematic error of ~2% in the n_o and n_e values due to uncertainty in the film thickness. The average difference $n_e - n_o$ at long wavelengths is 0.04 [cf. the inset of Fig. 3(b)]. Our n_o values agree well with those in Refs. 19 and 20, and our $\varepsilon_{\infty\perp}$ values fit well with those in Refs. 21 and 22. Theoretical estimates of ε_∞ for both polarizations by Chen *et al.*[23] and by Karch *et al.*[24] are about 3% larger and 2% smaller, respectively, than our values. The values of ε_∞ cited for both polarizations in Refs. 16–18 are typically ~1% smaller than those of Karch *et al.*[24]

These results have implications for lasers if M-plane GaN active layers are used with in-plane compressive strain ϵ_{xx}, $\epsilon_{zz} \lesssim -0.2\%$. The larger separation between the top VB states lowers the density of states at the VB top and will lead to lower transparent carrier density. Furthermore, the gain for transitions involving the topmost $|X\rangle$-like VB (E_1 transition) will be dominant. Therefore, if the laser cavity direction **C** of an edge emitter is such that **C**∥**c** (**C**∥**z**), efficient transverse-electric (TE) mode lasing (with **E**∥**x**) can occur. However, if **C**⊥ **c** (**C**∥**x**), the device would not work efficiently, because radiation associated with recombination involving the $|X\rangle$-like VB will not propagate along the **x** direction. Although in this configuration TE-mode lasing (with **E**∥**z**) would still be possible because of transitions to the $|Z\rangle$-like VB (E_2 transition), it would require harder pumping to achieve transparency, because the lower energy $|X\rangle$-like VB will get filled with holes first. For TE modes with **E**∥**x** and **E**∥**z**, the wave guide properties will be determined by n_o and n_e, respectively. These predicted features are in contrast to a C-plane edge emitter, where the device characteristics are independent of the cleaved-cavity orientation. In the case of vertical-cavity surface-emitting lasers with such active layers, the output can be expected to be strongly polarized for **E**⊥ **c**.

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[1] V. Fiorentini, F. Bernardini, F. Della Sala, A. Di Carlo, and P. Lugli, Phys. Rev. B **60**, 8849 (1999); F. Bernardini, V. Fiorentini, and D. Vanderbilt, *ibid.* **63**, 193201 (2001).
[2] T. Deguchi, K. Sekiguchi, A. Nakamura, T. Sota, R. Matsuo, S. Chichibu, and S. Nakamura, Jpn. J. Appl. Phys., Part 2 **38**, L914 (1999).
[3] P. Waltereit, O. Brandt, A. Trampert, H. T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, and K. H. Ploog, Nature (London) **406**, 865 (2000).
[4] P. Waltereit, O. Brandt, M. Ramsteiner, R. Uecker, P. Reiche, and K. H. Ploog, J. Cryst. Growth **218**, 143 (2000).
[5] M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1965).
[6] *Landolt-Börnstein, Series III*, edited by R. Allmann, W. Pies, and W. Weiss (Springer, Berlin, 1980), Vol. 7d2.
[7] M. Suzuki, T. Uenoyama, and A. Yanase, Phys. Rev. B **52**, 8132 (1995).
[8] A. J. Fischer, W. Shan, J. J. Song, Y. C. Chang, R. Horning, and B. Goldenberg, Appl. Phys. Lett. **71**, 1981 (1997).
[9] A. Shikanai, T. Azuhata, T. Sota, S. Chichibu, A. Kuramata, K. Horino, and S. Nakamura, J. Appl. Phys. **81**, 417 (1997).
[10] K. Domen, K. Horino, A. Kuramata, and T. Tanahashi, Appl. Phys. Lett. **70**, 987 (1997).
[11] The influence of strain on CB and VB states at the BZC was calculated separately. For the VB states, the Bir–Pikus (see Ref. 25) Hamiltonian (6×6 matrix) was solved. The deformation potentials (using the quasicubic approximation) were $D^{CB} = -44.5$ eV, $D_1^{VB} = -41.4$ eV, $D_2^{VB} = -33.3$ eV, $D_3^{VB} = 8.2$ eV, $D_4^{VB} = -4.1$ eV, and $D_5^{VB} = -4.7$ eV. Other strain-free EBS parameters were taken from Ref. 9.
[12] For the M-plane film under the constraint that the stress component $\sigma_{yy} = 0$, we obtain $\varepsilon_{yy} = -(C_{12}/C_{11})\varepsilon_{xx} - (C_{13}/C_{11})\varepsilon_{zz}$. C_{ij} denote the elastic constants (after Ref. 26).
[13] T. Ohtoshi, A. Niwa, and T. Kuroda, J. Appl. Phys. **82**, 1518 (1997).
[14] K. Domen, K. Horino, A. Kuramata, and T. Tanahashi, Appl. Phys. Lett. **71**, 1996 (1997).
[15] H. Y. Zhang, X. H. He, Y. H. Shih, M. Schurman, Z. C. Feng, and R. A. Stall, Appl. Phys. Lett. **69**, 2953 (1996).
[16] G. Yu, H. Ishikawa, T. Egawa, T. Soga, J. Watanabe, T. Jimbo, and M. Umeno, Jpn. J. Appl. Phys., Part 2 **36**, L1029 (1997).
[17] M. J. Bergmann, U. Özgür, H. C. Casey, Jr., H. O. Everitt, and J. F. Muth, Appl. Phys. Lett. **75**, 67 (1999).
[18] R. Goldhahn, S. Shokhovets, J. Scheiner, G. Gobsch, T. S. Cheng, C. T. Foxon, U. Kaiser, G. D. Kipshidze, and W. Richter, Phys. Status Solidi A **177**, 107 (2000).
[19] H. Amano, N. Watanabe, N. Koide, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 **32**, L1000 (1993).
[20] U. Tisch, B. Meyler, O. Katz, E. Finkman, and J. Salzman, J. Appl. Phys. **89**, 2676 (2001).
[21] A. S. Barker and M. Ilegems, Phys. Rev. B **7**, 743 (1973).
[22] T. Kawashima, H. Yoshikawa, S. Adachi, S. Fuke, and K. Ohtsuka, J. Appl. Phys. **82**, 3528 (1997).
[23] J. Chen, Z. H. Levine, and J. W. Wilkins, Appl. Phys. Lett. **66**, 1129 (1995).
[24] K. Karch, J.-M. Wagner, and F. Bechstedt, Phys. Rev. B **57**, 7043 (1998).
[25] G. L. Bir and G. E. Pikus, *Symmetry and Strain Induced Eeffects in Semiconductors* (Wiley, New York, 1974).
[26] A. Polian, M. Grimsditch, and I. Grzegory, J. Appl. Phys. **79**, 3343 (1996).