

Polarization anisotropy of the photoluminescence of *M*-plane (In,Ga)N/GaN multiple quantum wells

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We investigate the polarization anisotropy of the photoluminescence of an *M*-plane (1 $\bar{1}$ 00) In_{0.1}Ga_{0.9}N/GaN multiple quantum well grown on γ -LiAlO₂ (100) by molecular-beam epitaxy. In contrast to *C*-plane (0001) structures, a strong in-plane optical anisotropy with an energy-dependent polarization degree of up to 96% is observed for this *M*-plane sample. An apparent spectral shift of the emission with polarization angle is attributed to the impact of exciton localization on the polarization degree. The presence of localized states manifests itself further in the anomalous temperature dependence of the photoluminescence linewidth. © 2003 American Institute of Physics. [DOI: 10.1063/1.1579563]

Due to their hexagonal symmetry, wurtzite GaN and related compounds have a distinguished axis ([0001], the *c*-axis) and are thus inherently anisotropic materials. This anisotropy manifests itself in the spontaneous electric polarization along the polar *c*-axis, which is the common growth direction of these materials. This electric polarization is responsible for the large internal electrostatic fields within thin layers, such as quantum wells (QWs). Further consequences of the symmetry of GaN are the splitting of the valence band, different refractive indices for light parallel and normal to the *c*-axis, and an optically polarized spontaneous emission^{1–4} and absorption,⁵ even in the bulk.

It is clear from these considerations that the orientation of a GaN film has a decisive influence on its physical properties. Most prominently, heterostructures grown along directions perpendicular to the *c*-axis, such as [1 $\bar{1}$ 00] (*M*-plane)^{6,7} or [11 $\bar{2}$ 0] (*A*-plane),^{8,9} are free of the internal electrostatic fields along the growth direction. Additionally, a theoretical study using the *sp*³-tight-binding method implies that *M*-plane GaN/(Al,Ga)N QWs have a lower valence-band density of states and larger optical matrix elements than corresponding *C*-plane structures, resulting in improved lasing performance.¹⁰ Moreover, it has been recently shown that *M*-plane GaN is an attractive candidate for the realization of polarization sensitive detectors.¹¹ In this context, it is imperative to understand the selection rules for interband transitions in these structures. In previous work, we have addressed this issue by investigating the polarization anisotropy in *M*-plane GaN films⁵ and GaN/(Al,Ga)N multiple quantum wells¹² (MQWs). In agreement with theoretical predictions, the linear polarization degree of these structures is close to unity. An open and important question, however, is whether or not the ubiquitous exciton localization in (In,Ga)N destroys this strong polarization anisotropy. The recent realization of *M*-plane (In,Ga)N/GaN MQWs¹³ allows us to answer this question.

In this letter, we study the in-plane polarization anisotropy of the spontaneous emission of an *M*-plane (In,Ga)N/

GaN MQW by photoluminescence (PL). The polarization degree is found to be as high as 96%; that is, even higher than in GaN/(Al,Ga)N MQWs studied previously.¹² This finding presumably results from the large valence-band splitting induced by the high compressive strain in the (In,Ga)N wells. The lower polarization degree at the low-energy tail of the PL spectrum is the consequence of localization and causes an apparent shift of the spectrum with polarization angle.

The investigated sample was grown on γ -LiAlO₂(100) by plasma-assisted molecular-beam epitaxy and consists of 20 periods of 1.9-nm In_{0.1}Ga_{0.9}N and 6.6-nm GaN. Details concerning the growth conditions can be found in Ref. 13. Symmetric ω -2 θ triple-axis x-ray diffraction scans¹³ and resonant Raman scattering (not shown here) reveal a high phase-purity and the expected in-plane orientation relationship; that is, [0001]_{(In,Ga)N}||[010]_{LiAlO₂} and [11 $\bar{2}$ 0]_{(In,Ga)N}||[001]_{LiAlO₂}. The x-ray measurements, furthermore, allowed us to deduce that the In_{0.1}Ga_{0.9}N layers are subject to a biaxial compressive strain with an out-of-plane dilatation of 1.26%. Note that the in-plane strain is inevitably anisotropic, since both the lattice mismatch and the thermal expansion coefficients along the [0001] and [11 $\bar{2}$ 0] are not equal. The polarization anisotropy of the PL spectra was measured in backscattering geometry ($\mathbf{k} \perp c$) using a μ -PL setup, utilizing the 325-nm line of a He-Cd laser for excitation, and a liquid-N₂-cooled CCD camera for detection. The linear polarization of the PL at 10 K was analyzed by rotating a polarizer between a polarization angle ϕ of 0° and 360° (0° refers to $\mathbf{E} \parallel c$) in front of a single monochromator. The experimental data were carefully corrected for the polarization dependence of the setup, as confirmed by the fact that the luminescence of *C*-plane structures with similar emission energy is unpolarized, as theoretically expected. Temperature-dependent PL measurements were performed in a conventional setup using the same excitation and detection scheme as described earlier.

Figure 1 shows the low-temperature PL spectra as a function of ϕ ranging from 0° to 90°. The dominant PL band at 3.14 eV stems from the In_{0.1}Ga_{0.9}N/GaN MQW, while the higher-energy shoulder located near 3.36 eV origi-

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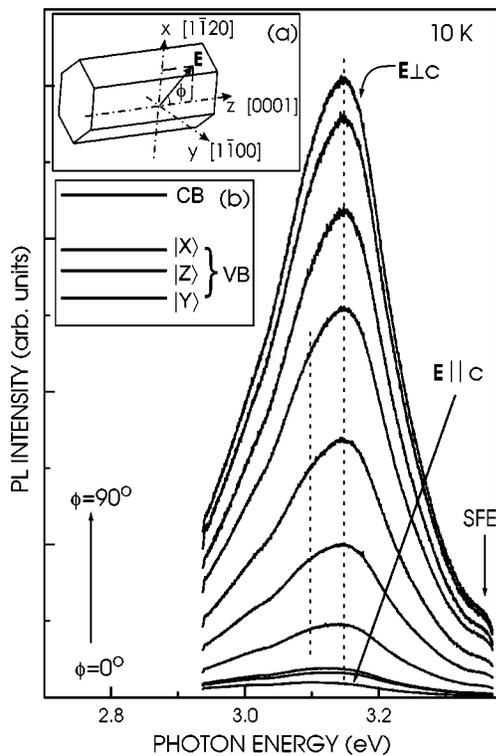


FIG. 1. μ -PL spectra of the M -plane $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}/\text{GaN}$ MQW at 10 K as a function of polarization angle from 0° to 90° . The dotted lines indicate the PL peak positions for $\mathbf{E}\perp c$ and $\mathbf{E}\parallel c$. Inset (a) shows a scheme of the wurtzite GaN unit cell and the coordinates used with the corresponding azimuths, whereas inset (b) illustrates schematically the valence-band order in strained M -plane GaN.

nates from excitons bound to stacking faults.¹⁴ Obviously, the entire emission is strongly polarized. However, contrary to the case of $\text{GaN}/(\text{Al,Ga})\text{N}$ MQWs,¹² the peak energy of the PL band varies with polarization angle. A low-energy shoulder located at around 3.1 eV becomes more prominent with decreasing polarization angle, and eventually dominates the spectrum for $\phi=0^\circ$; that is, $\mathbf{E}\parallel c$.

This finding is surprising, since it seems at first glance to contradict our understanding of the bandstructure of M -plane GaN. As described in detail in Ref. 15, the anisotropic in-plane compressive strain experienced by the $(\text{In,Ga})\text{N}$ films breaks the symmetry in the x - y plane of the wurtzite crystal and renders the original $|X \pm iY\rangle$ -like valence-band states of unstrained wurtzite GaN into $|X\rangle$ -like and $|Y\rangle$ -like states (see Fig. 1 for our choice of coordinates). The $|X\rangle$ -like state is raised in energy by the strain, while the $|Y\rangle$ -like state is pushed down below the $|Z\rangle$ -like state (cf., Fig. 1). The inter-band transition lowest in energy is thus that involving the $|X\rangle$ -like valence band which, according to its symmetry, is expected to be predominantly x -polarized ($\mathbf{E}\perp c$). The transition involving the $|Z\rangle$ -like valence bands occurs at higher energy and is expected to be z -polarized ($\mathbf{E}\parallel c$). In our experiment, however, the situation seems reversed, as the PL band blueshifts when going from $\mathbf{E}\parallel c$ to $\mathbf{E}\perp c$.

To resolve this unexpected shift of the PL band, we next examine the PL spectra in more detail. Figure 2 presents the normalized PL spectra for $\phi=0^\circ$ and 90° as well as the energy-resolved polarization degree. The spectrum taken with $\mathbf{E}\perp c$ peaks at 3.14 eV, but exhibits a (poorly resolved) shoulder at 3.1 eV, just where the spectrum taken with $\mathbf{E}\parallel c$

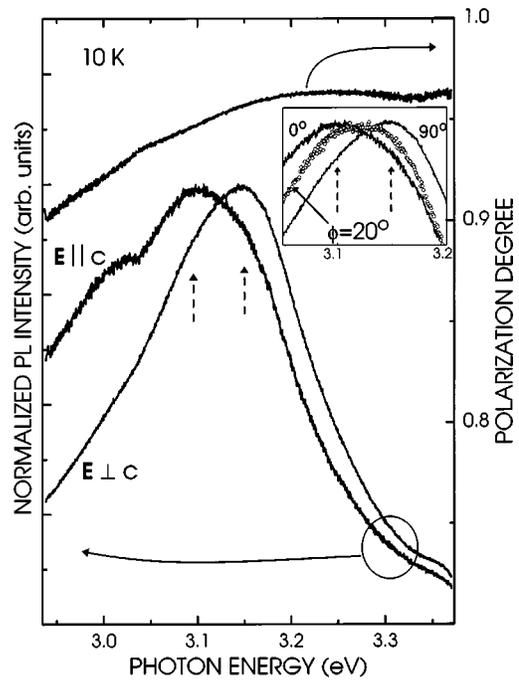


FIG. 2. Normalized μ -PL spectra polarized parallel and perpendicular to the c -axis for the M -plane $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}/\text{GaN}$ MQW. The polarization degree as a function of photon energy is also shown. The arrows indicate the positions for the PL maximum in one spectrum and the corresponding shoulder in the other one. The inset is a magnified view of the normalized spectra at $\phi=0^\circ, 20^\circ$, and 90° .

has its maximum. Similarly, the latter spectrum exhibits a bump at 3.14 eV, corresponding to the maximum of the former (the shoulder at 3.01 eV might be due to a phonon replica of the dominant transition in this spectrum). It thus seems that the PL band consists of (at least) two contributions, the respective strength of which varies with polarization angle. Indeed, as shown in the inset of Fig. 2, the spectrum taken at $\phi=20^\circ$ exhibits a top-hat shape, indicating that these two contributions have equal strength at this polarization angle. We attribute this finding to the superposition of transitions involving extended states and a band of localized states. This interpretation is confirmed by the energy dependence of the polarization degree ρ , obtained in the usual way, as

$$\rho = \frac{I_\perp - I_\parallel}{I_\perp + I_\parallel}, \quad (1)$$

where I_\perp (I_\parallel) is the PL intensity for $\mathbf{E}\perp c$ ($\mathbf{E}\parallel c$). As seen in Fig. 2, the polarization degree thus obtained amounts to 96% at high energies, but gradually decreases to 90% with decreasing energy. The degree of polarization is thus even higher than that found for $\text{GaN}/(\text{Al,Ga})\text{N}$ MQWs, which is likely to be a result of the large valence-band splitting induced by the high compressive strain within the wells.¹² The monotonic decline of ρ with decreasing emission energy has been observed previously for excitons localized by compositional fluctuations in $\text{CdS}_{1-x}\text{Se}_x$.¹⁶ A possible explanation for this behavior is the increased mixing of the different valence-band states due to stronger confinement, which destroys their unique symmetry. Ultimately, a localized exciton approaches a zero-dimensional entity with a “symmetrized” wave function and an entirely unpolarized emission.^{17,18} In

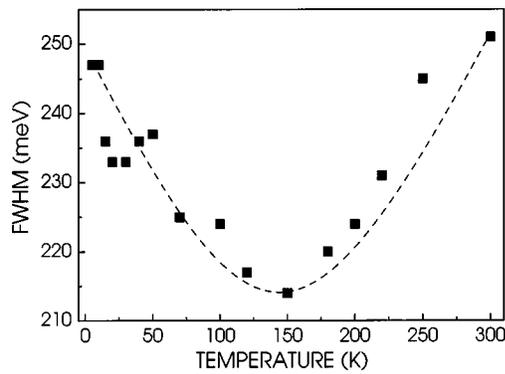


FIG. 3. FWHM of the PL spectrum as a function of temperature. The dashed line is a guide for the eye.

any case, it is clear that the spectral shift of the PL band is an apparent one, in that it results solely from the different ρ of the states involved in the emission process. The states involved in the emission process are of predominant $|X\rangle$ -like character. Even considering the high strain within the wells, the valence-band splitting is unlikely to exceed the total width of the PL band. This finding thus implies that the PL band stems almost entirely from transitions involving localized states of different potential depths that are not in equilibrium with each other.¹²

Finally, the actual presence of localized states is confirmed by the anomalous temperature dependence of the full width at half-maximum (FWHM) of the PL spectrum, as shown in Fig. 3. The narrowing of the spectrum upon increasing temperature has been observed in several materials systems in which strong localization occurs, such as InAs/GaAs quantum dots,¹⁹ (In,Ga)As/GaAs QWs,²⁰ and (In,Ga)N QWs.²¹ At low temperatures, excitons are randomly trapped by the localizing centers, and the PL linewidth basically corresponds to the width of the distribution of local potential minima. With increasing temperature, excitons are thermally activated and become mobile within the band of localized states, allowing them to relax further into deeper potential minima. This thermal activation will thus result in the distinct narrowing of the linewidth. At even higher temperature, the thermal distribution of excitons will force them to populate higher-energy states as well. As a result, the PL band broadens and its width once again approaches the bandwidth. Note that the notion of a band of localized states is also required for the understanding of the recombination dynamics observed for this structure, which exhibits a distinct spectral diffusion upon pulsed excitation.¹³

In summary, a strong polarization anisotropy with a linear polarization degree of up to 96% has been observed for an M -plane $\text{In}_{0.1}\text{Ga}_{0.9}\text{N}/\text{GaN}$ MQW. This very high degree of polarization is presumably related to a large valence-band splitting induced by the large compressive strain within the wells. The apparent spectral shift of the PL band with polarization angle was shown to be due to the progressively lower polarization degree of localized states with increasing localization depth. Localized states in nonpolar (In,Ga)N/GaN MQWs thus not only govern the recombination dynamics in this material, but also have an important impact on the polarization anisotropy.

- ¹A. Niwa, T. Ohtoshi, and T. Kuroda, *Jpn. J. Appl. Phys., Part 2* **35**, L599 (1996).
- ²M. Suzuki and T. Uenoyama, *Jpn. J. Appl. Phys., Part 1* **35**, 543 (1996).
- ³R. Dingle, D. D. Sell, S. E. Stokowski, and M. Ilegems, *Phys. Rev. B* **4**, 1211 (1971).
- ⁴K. Domen, K. Horino, A. Kuramata, and T. Tanahashi, *Appl. Phys. Lett.* **71**, 1996 (1997).
- ⁵S. Ghosh, P. Waltereit, O. Brandt, H. T. Grahn, and K. H. Ploog, *Appl. Phys. Lett.* **80**, 413 (2002).
- ⁶P. Waltereit, O. Brandt, A. Trampert, H. T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, and K. H. Ploog, *Nature (London)* **406**, 865 (2000).
- ⁷E. Kuokstis, C. Q. Chen, M. E. Gaeovski, W. H. Sun, J. W. Yang, G. Simin, M. A. Khan, H. P. Maruska, D. W. Hill, M. C. Chou, J. J. Gallagher, and B. Chai, *Appl. Phys. Lett.* **81**, 4130 (2002).
- ⁸H. M. Ng, *Appl. Phys. Lett.* **80**, 4369 (2002).
- ⁹M. D. Craven, S. H. Lim, F. Wu, J. S. Speck, and S. P. DenBaars, *Appl. Phys. Lett.* **81**, 469 (2002).
- ¹⁰A. Niwa, T. Ohtoshi, and T. Kuroda, *Appl. Phys. Lett.* **70**, 2159 (1997).
- ¹¹S. Ghosh, O. Brandt, H. T. Grahn, and K. H. Ploog, *Appl. Phys. Lett.* **81**, 3380 (2002).
- ¹²B. Rau, P. Waltereit, O. Brandt, M. Ramsteiner, K. H. Ploog, J. Puls, and F. Henneberger, *Appl. Phys. Lett.* **77**, 3343 (2000).
- ¹³Y. J. Sun, O. Brandt, S. Cronenberg, S. Dahr, H. T. Grahn, K. H. Ploog, P. Waltereit, and J. S. Speck, *Phys. Rev. B* **67**, 041306(R) (2003).
- ¹⁴Y. J. Sun, O. Brandt, U. Jahn, T. Y. Liu, A. Trampert, S. Cronenberg, S. Dhar, and K. H. Ploog, *J. Appl. Phys.* **92**, 5714 (2002).
- ¹⁵S. Ghosh, P. Waltereit, O. Brandt, H. T. Grahn, and K. H. Ploog, *Phys. Rev. B* **65**, 075202 (2002).
- ¹⁶S. Y. Verbin, S. A. Permogorov, and A. N. Reznitskiĭ, *Sov. Phys. Solid State* **25**, 195 (1983).
- ¹⁷F. Vouilloz, D. Y. Oberli, M.-A. Dupertuis, A. Gustafsson, F. Reinhardt, and E. Kapon, *Phys. Rev. B* **57**, 12378 (1998).
- ¹⁸A. Crottini, J. Staehli, B. Deveaud, X.-L. Wang, and M. Ogura, *Phys. Rev. B* **63**, 121313(R) (2001).
- ¹⁹A. Patanè, M. G. Alessi, F. Intonti, A. Polimeni, M. Capizzi, F. Martelli, M. Geddo, A. Bosacchi, and S. Franchi, *Phys. Status Solidi A* **164**, 493 (1997).
- ²⁰M. G. Alessi, F. Fragano, A. Patanè, M. Capizzi, E. Runge, and R. Zimmermann, *Phys. Rev. B* **61**, 10985 (2000).
- ²¹P. Lefebvre, T. Talierno, A. Morel, J. Allègre, M. Gallart, B. Gil, H. Mathieu, B. Damilano, N. Grandjean, and J. Massies, *Appl. Phys. Lett.* **78**, 1538 (2001).