Nonpolar and Semipolar Group III Nitride-Based Materials

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Abstract

GaN and its alloys with InN and AIN are materials systems that have enabled the revolution in solid-state lighting and high-power/high-frequency electronics. GaN-based materials naturally form in a hexagonal wurtzite structure and are naturally grown in a (0001) c-axis orientation. Because the wurtzite structure is polar, GaN-based heterostructures have large internal electric fields due to discontinuities in spontaneous and piezoelectric polarization. For optoelectronic devices, such as light-emitting diodes and laser diodes, the internal electric field is generally deleterious as it causes a spatial separation of electron and hole wave functions in the quantum wells, which, in turn, likely decreases efficiency. Growth of GaN-based heterostructures in alternative orientations, which have reduced (semipolar orientations) or no polarization (nonpolar) in the growth direction, has been a major area of research in recent years. This issue highlights many of the key developments in nonpolar and semipolar nitride materials and devices.

Background

The Group III nitrides are a remarkable materials system. With direct bandgaps ranging from 0.7 eV (InN) through 3.4 eV (GaN) to 6.0 eV (AlN), this materials system has enabled deep ultraviolet (λ < ~300 nm or photon energy > ~4.1 eV based on high Al content Al, Ga1-xN quantum wells [QWs]), ultraviolet ($\lambda < \sim 400$ nm or photon energy > \sim 3.1 eV), blue ($\lambda \approx 455$ nm or photon energy = 2.7 eV based on $In_{\nu}Ga_{1-\nu}N$ QWs), and green ($\lambda = 525$ nm or photon energy = 2.4 eV) emitters based on In, Ga1-, N QWs, and longer wavelength light-emitting diodes (LEDs) and violet and blue laser diodes (LDs). No other materials system offers this range of direct bandgaps. Prior to the development of the nitrides as optoelectronic materials, there were no efficient ultraviolet or blue LEDs, and green LEDs based on II-VI materials had poor lifetimes due to the ease of dislocation generation in the active regions of the II-VIs. Devices based on Group III nitrides perform well despite threading dislocation densities typically in excess of 108 cm-2 (but poorly when the dislocation density exceeds 1010 cm-2, thus pushing the drive for bulk GaN substrates as described by Fujito et al.'s article in this issue) in comparison to conventional III-V materials such as GaAs, where the dislocation densities are typically 104 cm-2 or less. However, when the nitrides are remarkably robust against oxidation (and etching), they are mechanically hard and show no evidence for extended defect motion at standard operating temperatures. GaNbased LEDs feature prominently in colored lighting applications, such as green traffic lights. When blue LEDs are combined with a yellow-emitting phosphor, white light is produced; thus, GaN has been the material that enabled the field of solid-state lighting. Violet GaN-based LDs operating at a wavelength of 405 nm are already in broad use in high definition optical data storage.

The articles in this issue describe recent progress in materials and device developments for Group III-based materials. The use of new crystallographic orientations for GaN and AlN may enable major performance improvements over/in existing device technologies and new device functionalities (see, for instance, the introduction to the article by Suda and Horita on the potential for high-efficiency deep ultraviolet emitters based on nonpolar AlN).

Starting with the pioneering work of Maruska et al. on GaN in the late 1960s and early 1970s with hydride vapor-phase epitaxial (HVPE) growth1,2 and continuing with the subsequent major breakthroughs by Amano et al. in the 1980s on two-step growth and p-type GaN activation3,4 and then by Nakamura et al. in the early to mid 1990s,5-9 nearly all work on GaN-based materials and devices has been on c-plane (0001)-oriented layers. Due to the inability to fabricate bulk GaN crystals (due to its high melting point and vapor pressure) at the time of early GaN pioneering work, an area that still remains a major challenge for nitride research, GaN-based technology, has been developed on non-native, or foreign, substrates. The highest quality GaN has been grown in the (0001) orientation on either (0001)oriented sapphire or (0001)-oriented 4Hor 6H-SiC

Through the mid-to-late 1990s, there was a broadening realization of the importance of piezoelectricity and polarization in Group III nitrides.10-13 Since the wurtzite structure (hexagonal; space group P63mc; point group 6mm-see Figure 1) is noncentrosymmetric (absence of inversion symmetry), it is polar, with spontaneous polarization occurring parallel to the -c-axis. Polarization-related effects are a dominant feature in common c-axis GaN-based heterostructures and a major differentiating feature of the wurtzite Group III nitrides from other well-developed semiconductors, such as silicon or gallium arsenide.

Polarization in the Group III Nitrides

At the same time that Nakamura was making major breakthroughs in GaNbased materials and devices, Resta14 and Vanderbilt and Kingsmith^{15,16} were developing the quantum theory of polarization. In their landmark work, Vanderbilt and Kingsmith showed that polarization in a solid is a bulk property and can be determined quantum mechanically with knowledge of the phase of the valence electron wave functions (for a review of these developments, see References 17 and 18). Subsequently, Bernardini et al.19 calculated the spontaneous polarization and piezoelectric constants for GaN, AlN, and InN; this work is the standard used today for these important physical properties in Group III nitrides.

The spontaneous polarization P_o values for nitrides are as high as ~30% of the values for classical proper ferroelectrics such as BaTiO₃ ($P_s = 0.26 \text{ C/m}^2$) (i.e., $P_s =$ -0.081 C/m², -0.029 C/m², -0.032 C/m² for AlN, GaN, and InN, respectively). Within the Group III nitrides, they have piezoelectric coefficients with similar magnitudes and the same sign. Note in Figure 1 that the sense of the spontaneous polarization is parallel to the [0001] direction for GaN and similarly for AlN and InN; this means that the metal-terminated (0001) surface has a negative fixed polarization sheet charge. The discontinuity in total polarization in (0001)-oriented nitride-based interfaces in heterostructures results in significant fixed charges at the interfaces that may be beneficial or deleterious depending on the final device technology.

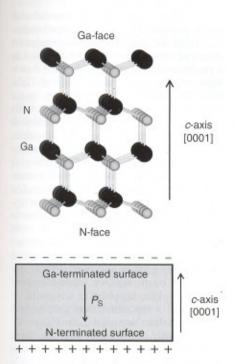


Figure 1. Ball-and-stick model of the wurtzite GaN crystal structure (top schematic). The gallium atoms are represented as large darker spheres, and the nitrogen atoms are small lighter spheres. The top crystal face (0001) also is referred to as the gallium face. This is the orientation for nearly all commercial GaN-based devices. The bottom schematic depicts a slab of GaN with the (0001) surface at the top showing negative fixed polarization charges, and the bottom is showing a (000T) nitrogen face with positive fixed polarization charge.

For example, the alloying of GaN can be taken with the wider bandgap semiconductor AlN to form Al, Ga,N, which has a smaller relaxed lattice constant than GaN, and coherently grow the Al, Ga, N alloy on a (0001)-oriented GaN layer. Since the underlying GaN structure is nominally relaxed and the Al, Ga,N layer is under an equibiaxial tensile stress, there will be additional piezoelectric polarization in the Al, Ga, N layer. The sense and magnitude of the spontaneous polarization discontinuity between GaN and Al, Ga,N are similar to the piezoelectric polarization in the coherent Al, Ga, N layer. Thus, the polarization discontinuity at the Al, Ga, N/GaN interface gives rise to a significant positive fixed sheet charge that may be balanced by free electrons in a two-dimensional electron gas (2DEG) when the AlGaN layer has a suitable thickness, as shown schematically in Figure 2. The 2DEG has a very narrow width, on the order of 2-3 nm, in the direction perpendicular to the interface and holds a high density of mobile free electrons. For commonly employed Al_{0.30}Ga_{0.70}N/GaN high electron mobility transistor structures, the electron concentration in the 2DEG is on the order of 1013 cm-2. For electronic devices, polarization is largely seen as beneficial as it gives rise to 2DEG sheet concentrations nearly an

order of magnitude larger than those in conventional III–V systems. The 2DEG high electron concentration, combined with high saturation velocity and high breakdown field, has made the nitrides the preferred materials for high-power and high-frequency electronics applications (see References 20 and 21 for a brief and more detailed overview of GaN-based electronics).

For LEDs (e.g., based on GaN barriers and In, Ga1-x N QWs), the fixed sheet charges at the heterointerfaces cause large electric fields normal to the QW plane (fields on the order of 1 to 2 MV/cm depending on the InN molar fraction x) that are in the opposite sense to the polarization-related electric fields in the Al, Ga, N/GaN example shown in Figure The difference in spontaneous polarization between In, Ga, N and GaN is small, but the In, Ga1-, N layers are coherently strained (i.e., under equibiaxial compressive stress to the GaN buffer and barriers), and large internal electric fields result in c-plane In, Ga1-xN QWs, as shown in the band profiles in Figure 3a, which were produced by self-consistent solutions to the Schrödinger and Poisson equations. In optoelectronic devices such as LEDs, the In, Ga1-, N QWs are placed in the depletion region of a p-n diode such that injected electrons and holes are captured

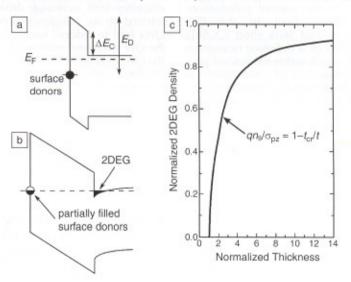


Figure 2. Schematic band diagram for an $AI_xGa_{1...x}N/GaN$ heterostructure used to form the two-dimensional electron gas (2DEG) for GaN-based transistors. (a) Band profile for a thin $AI_xGa_{1...x}N$ layer on GaN. The electric field in the AIGaN layer (slope of bands) is due to discontinuities in spontaneous polarization. (b) Band profile for a thicker $AI_xGa_{1...x}N$ layer on GaN, where surface donors are responsible for charge transfer from the surface to the back $AI_xGa_{1...x}N/GaN$ interface to form the 2DEG. (c) Predicted 2DEG sheet charge density from the surface donor model. (For more details, see Reference 43.) In the figures, E_F is the Fermi level; ΔE_C is the conduction band offset; E_D is the surface donor energy level; q is the fundamental charge; n_x is the 2DEG sheet charge density; σ_{po} is the fixed polarization charge; t is the $AI_xGa_{1...x}N$ layer thickness; and t_{cr} is the critical layer thickness to form the 2DEG.

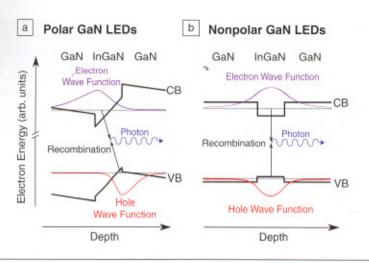


Figure 3. Self-consistent Schrödinger-Poisson solutions for the band profile (conduction band [CB] and valence band [VB]) for GaN/In_{0.10}Ga_{0.90}N(25Å)/GaN quantum well (QW) for the conventional polar (0001) Ga-face orientation. (a) This is typically used in violet GaN-based light-emitting diodes (LEDs) and laser diodes and for (b) a nonpolar orientation corresponding to either *a*-plane or *m*-plane orientation. Note the large electric field in the conventional polar orientation, which results in a large spatial separation of the electron and hole wave functions. In contrast, the ground-state electron and hole wave functions directly overlap in the nonpolar InGaN QW.

by the QW without recombination in the surrounding material. These internal polarization-related electric fields cause spatial separation of electron and hole wave functions. When the electron and hole wave functions are spatially separated due to the internal polarizationrelated electric field in the QW (quantum-confined Stark effect [QCSE]), the likelihood of a radiative recombination event, which yields the desired photon, is reduced, and thus, there is increased likelihood of the injected electrons or holes to nonradiatively recombine and the energy is squandered as heat. Thus, the polarization-related electric fields are seen to reduce efficiency in nitride LEDs and LDs. To avoid the electric field-induced electron-hole wave function separation, the thickness of In, Ga1-rN QWs is limited to ~25 Å as a balance between tolerable confinement of injected carriers and the separation of electron and hole wave functions in nearly all commercial GaN-based LEDs or LDs. Other key features of c-plane GaN-based LEDs include a significant blueshift in the electroluminescence (EL) and an efficiency droop with increasing drive current. The EL blueshift is caused by the combined effects of Coulomb screening of the internal electric field mentioned earlier and band filling of the tail states by the increase in carrier concentration in the QWs.11 Because the growth of perfectly homogeneous In, Ga1-xN alloys is difficult due to the thermodynamic mismatch between GaN and InN, In, Ga1-xN alloys

have pronounced tail states in their band edge. The difficulty in growing atomically flat In, Ga1., N QWs at low growth temperature also gives rise to the energy tail states because of a thickness fluctuation in the QWs. With respect to decreased device efficiency with increasing drive current, referred to as "efficiency droop," thin QWs lead to reduced barrier heights for the QW states and reduced total states in the QW, thus increasing the likelihood of electron overflow. In other words, at high current injection and a higher total carrier density, reduced barrier height and available states can cause an efficiency droop through the decreased injection efficiency (i.e., injected carriers overflowing from the QW) and an increase in nonradiative processes such as Auger recombination (see Reference 22 for more on the evidence for Auger processes in InGaN).

Nonpolar Orientations

The challenges associated with polar heterostructures in the nitrides could be avoided entirely by growth in nonpolar orientations (i.e., with the c-axis of the wurtzite structure parallel to any heterojunction). Due to the crystal symmetry, there would be no polarization discontinuity at the heterointerface. The low index a-plane {1120} and m-plane {1100} are candidate orientations for fabricating nonpolar heterostructures. Through the 1990s, there were scattered reports of nonpolar GaN growth, but the films typically had rough three-dimensional morphologies which consisted of exposed semipolar [1011] and

N-face (000T) facets, and in the context of polarization, there were few, if any, reports of wurtzite GaN growth in nonbasal plane orientations to avoid polarization-related electric fields.^{23–30} By 2000, there was growing recognition that nonpolar orientations and alternative orientations, with reduced polarization discontinuities at heterointerfaces, could yield devices with higher optical transition probabilities due to the increased overlap of the electron and hole wave functions, as shown in Figure 3b for nonpolar In, Ga_{1-x}N QWs.³¹

In 2000, Waltereit et al. in Klaus Ploog's group at the Paul Drude Institute in Berlin reported on the first planar nonpolar (1100) m-plane films on (100)-oriented γ-LiAlO₂ substrates.³² In their breakthrough article, Waltereit et al. demonstrated the absence of internal electric fields in m-plane GaN QWs with AlGaN barriers.³² Waltereit's work motivated a worldwide research effort in nonpolar

and semipolar nitrides. γ-LiAlO₂ is an interesting material and can be produced in relatively large boules. For epitaxial growth by metalorganic chemical vapor deposition (MOCVD), it is a problematic substrate because it has a high sublimation rate at typical GaN MOCVD growth temperatures (e.g., 1100°C). γ-LiAlO₂ readily reacts with moisture in the air, and this has presented practical problems in its use as a substrate. In 2001–2002, researchers at the University of California, Santa Barbara (UCSB) and elsewhere learned that Group III nitride growth on r-plane (10 $\overline{12}$) sapphire yields a-plane (1120) films with only one film orientation; however, previous growth efforts always yielded rough films with faceted three-dimensional morphologies.33,34 Craven et al.33 discovered by MOCVD growth and Ng23 realized by molecular beam epitaxy planar a-plane films on r-plane sapphire (a schematic of the typical x-ray diffraction results and film orientation are shown in Figure 4). Until recently, the r-plane was the most common orientation for sapphire substrates, and thus Craven's and Ng's results provided a pathway to large-area low-cost nonpolar films.

The absence of the polarization field effects in nonpolar QWs was confirmed by the distinctly short radiative lifetime of *m*-plane AlGaN/GaN QWs compared with the *c*-plane radiative lifetime because in the nonpolar QWs, the electron and hole wave functions have strong overlap, whereas in the *c*-plane QWs, the wave functions are spatially separated.³² At the same time, the cathodoluminescence peak energy of *m*-plane QW was higher than that of *c*-plane QW, which

was explained purely by the quantum size effect. After the finding, a similar absence of the QCSE has been reported for a-plane AlGaN/GaN QWs in a-plane GaN.^{34,35}

Semipolar Orientations

A significant effort in the same timeframe was focused on discovering a largearea low-cost foreign substrate that would yield m-plane GaN films, as a collective body of evidence has shown that the m-plane is the more stable plane in comparison to the a-plane. An important result from this search was the development of films with an orientation between nonpolar planes and polar c-planes. These film orientations were coined "semipolar" (i.e., semipolar planes are those with a nonzero h, k, or i index and a nonzero l index in the [hkil] Miller-Bravais indexing convention).36 Some common semipolar orientations are shown in Figure 5. Although polarization discontinuities in semipolar heterostructures are not entirely eliminated, they are significantly reduced, as first shown by Takeuchi et al.12,31 and more recently by Romanov et al.37 Figure 6 shows the total polarization discontinuity ΔP_z for $In_xGa_{1-x}N$ layers coherently strained to GaN for crystallographic orientations θ ranging from the polar c-plane $(\theta = 0^{\circ})$ to nonpolar $(\theta = 90^{\circ})$ for indium contents x typically used in violet to blue LEDs. There is a zero crossing in the total polarization discontinuity at ~45° orientation and significantly reduced polarization discontinuity at inclination angles of ~60°, which are close to two low-index semipolar orientations, (1011) and (1122) $(\theta = \sim 62^{\circ} \text{ and } \theta = \sim 58^{\circ}, \text{ respectively}), \text{ and }$ can be achieved as inclined sidewalls in selective area growth (see the articles in this issue by Funato and Kawakami and by Scholz et al.).

Early Nonpolar Devices

At the initial stage of the nonpolar/ semipolar nitride research, most of the epitaxial films and QWs were grown on heteroepitaxial substrates such as a-plane GaN on r-plane sapphire, as described earlier, m-plane GaN on (100) γ-LiAlO₂ or m-plane SiC, and semipolar GaN on m-plane sapphire or other substrates. With few or no exceptions, nonpolar or semipolar Group III nitride films grown on nonnative substrates have a high density of basal plane stacking faults (BPSFs), with a typical density in the range 105 cm-1 to 106 cm-1 in addition to a high density of threading dislocations (TDs)-typically in excess of 1010 cm-2,33 The SFs in nonpolar GaN films have been shown to emit characteristic photoluminescence peaks at 3.42

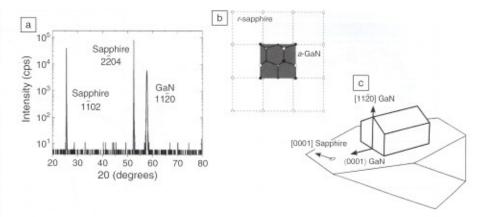


Figure 4. (1T02) r-plane sapphire is a widely available large-area high-quality substrate. Growth of wurtzite Group III nitrides on r-plane sapphire results in phase pure a-plane material, as shown in the example of the high-resolution x-ray diffraction of a-plane GaN. (a) High-resolution x-ray diffraction showing phase pure a-plane (11Z0) GaN on r-plane (1T02) sapphire. (b) Matching of the a-plane GaN lattice mesh with the r-plane sapphire lattice mesh. (c) Relationship between the crystal orientation of a-plane GaN and r-plane sapphire. (See Reference 33 for more details.)

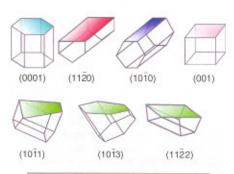


Figure 5. Common low-index orientations of wurtzite GaN (four-index notation) and cubic GaN (three-index notation). The figure shows the polar (0001) or c-plane; the nonpolar a-plane (1120) and m-plane (1010); the semipolar planes (1011), (1013), and (1122); and the nonpolar (001) plane for cubic GaN.

and 3.29 eV,38 which reduce the emission efficiency of the real band-edge related luminescence. Also, TDs and BPSFs may generate nonradiative recombination centers and thus reduce room temperature emission intensity. Figure 7a summarizes the dependence of the external quantum efficiencies (EQEs, which is the fraction of injected electron-hole pairs that form a photon that can escape from the semiconductor—the highest possible EQE is 100%) of nonpolar/semipolar QW LEDs on emission wavelength. For comparison, EQEs of state-of-the-art c-plane LEDs are also shown in Figure 7. The EQEs of defective nonpolar LEDs grown on foreign substrates (blue triangles) were far lower than c-plane devices and longer

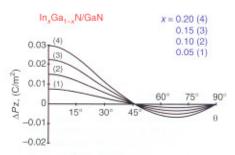
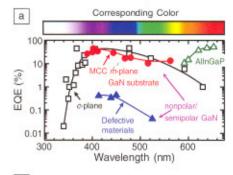


Figure 6. Dependence of the total polarization discontinuity on crystal orientation for a $\ln_x Ga_{1-x}N$ coherently strained to a thick GaN layer for In contents in InGaN from 5 to 20%. $\theta=0^\circ$ corresponds to the conventional c-plane orientation, and $\theta=90^\circ$ corresponds to nonpolar orientations. The total polarization discontinuity for InGaN on GaN is mostly due to strain-induced polarization (piezoelectric) in the compressively strained InGaN layer. Note the zero crossing in polarization discontinuity at -45° . (See Reference 37 for more details.)

wavelength LEDs based in the AlInGaP system.

Polarized Light Emission

In 2005, using defective *m*-plane GaN-based LEDs grown on *m*-plane SiC, Gardner et al. demonstrated another interesting feature of nonpolar structures—polarized light emission.³⁹ For GaN, the valence band (VB) has the character of nitrogen *p*-orbitals, and thus the three VBs have *x*, *y*, *z* symmetry corresponding to the nonpolar *a* and *m* directions and to the *c* direction, respectively. As shown in



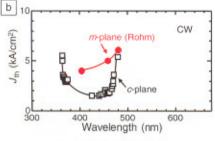


Figure 7. (a) Dependence of external quantum efficiency (EQE) on wavelength for conventional c-plane GaN lightemitting diodes (LEDs) (open squares); early nonpolar LEDs on Mitsubishi Chemical Cooperation (MCC) m-plane substrates (red circles); nonpolar LEDs grown on foreign substrates (blue triangles); and LEDs at longer wavelengths in the AllnGaP system. (Based on information from References 44-51; guidelines from References 52 and 53.) (b) Dependence of best reported threshold current density on lasing wavelength for nitride-based lasers. J_{th}, threshold current density; CW, continuous wave, (Based on information from References 54-61.)

Figure 8, the top two VBs (i.e., heavy hole and light hole VBs, respectively) have symmetry $|X+iY\rangle$ and $|X-iY\rangle$, and the split-off band has symmetry | Z). For c-plane structures, the dipole allowed transitions from the conduction band (which is primarily composed of Ga s-like states and is thus isotropic) to the top two VBs, which gives isotropic polarized direct light emission due to the six-fold symmetry of the c-axis orientation. The reduced symmetry (in comparison with c-plane oriented QWs) within the compressively stressed nonpolar In_{1-x}Ga_xN QW results in the possibility of direct emission of polarized light-primarily due to transitions from the conduction band to the top VB. The potential for engineering In_{1-x}Ga_xN QWs in nonpolar orientations for polarized light emission and detection has been an active area of research (see the articles by Funato and Kawakami and by Grahn in this issue, and see Reference 40).

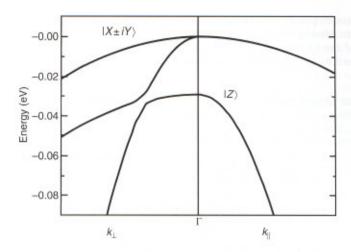


Figure 8. Schematic valence band structure for wurtzite GaN. The top two valence bands are nearly degenerate at the Γ point and have $|X\pm iY\rangle$ symmetry, which corresponds to the in-plane a or m directions. The lower valence band has predominantly $|Z\rangle$ symmetry, which corresponds to the [0001] orientation. Excitonic transitions for the conduction band to the three valence bands often are referred to as A, B, and C excitons. (Figure courtesy of P. Rinke and C.G. Van de Walle, University of California, Santa Barbara.) See Reference 62 for more information.

High-Performance Devices on High-Quality Substrates

During 2005-2006, there was a transformation in nonpolar and semipolar GaN research. Researchers at Furukawa Company and at Mitsubishi Chemical Corporation developed several millimeter thick c-plane GaN boules by HVPE (see the articles by Fujito et al. and by Funato and Kawakami in this issue, and see Reference 41). Subsequently, low defect density nonpolar and semipolar GaN substrates can be prepared by wafering these boules in a suitable orientation. Referring back to Figure 7a, the EQE values of m-plane devices (red circles) already rival those of the highly developed conventional c-plane devices. Also, the threshold current density of m-plane InGaN/GaN LDs grown on the m-plane GaN substrates (red circles) became comparable to that of c-plane LDs (open squares), especially for bluish green color regions, as shown in Figure 7b. The groups at Rohm and at UCSB have produced nonpolar and semipolar LEDs that rival the performance of state-of-the-art c-plane devices (see the articles by Funato and Kawakami and by Feezell et al. in this issue). The Rohm and UCSB groups demonstrated the first nonpolar LDs and the first AlGaN clad-free LDs as described in the article by Ohta and Okamoto and Reference 42. The promise of nonpolar light emitters has now been realized. In the coming years, we will learn whether nonpolar structures will be the ultimate solution for realizing nitride emitters at many different wavelengths and for a variety of electron devices.

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