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# Near-band-edge photoluminescence of wurtzite-type AlN

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Temperature-dependent photoluminescence (PL) measurements were performed for A-plane and C-plane bulk AlN single crystals and epitaxial layers on sapphire. A strong near-band-edge (NBE) emission and deep-level luminescence were observed. At low excitations, the emission spectra are dominated by free and bound excitonic transitions and their LO-phonon replicas. At high excitations, the broadening and redshift of the NBE band is attributed to dense electron–hole plasma formation. The PL spectra differences of bulk single crystals and epilayers is explained by the electron–hole plasma expansion peculiarities. *© 2002 American Institute of Physics.* [DOI: 10.1063/1.1510586]

Due to the potential use of III-nitride materials for both the blue-ultraviolet (UV) optoelectronic and microelectronics applications, they have been the subject of intense research efforts for more than a decade.[1,2] However, up to now the research focus has primarily been GaN, ternary InGaN, AlGaN, and quaternary AlInGaN alloys with optical transitions in the 280–550 nm range.[3–5] Molecular beam epitaxy or metalorganic chemical vapor deposition (MOCVD) techniques were primarily used to grow the studied high-quality films and quantum structures on substrates such as sapphire, SiC, or bulk GaN. To push the optical emission/detection wavelength to the deep UV region ($x \leq 280$ nm) high-quality $Al_xGa_{1-x}N$ alloys with Al-mole fractions in excess of $x = 0.5$ are needed. Deposition of these compounds on sapphire, SiC, or GaN is challenging due to a large lattice mismatch and thus, very small critical thickness of the layers. For such depositions, single-crystalline (SC) AlN can serve as an ideal substrate. In addition, high-optical-quality AlN layers are an excellent choice for the cladding region for heterostructure devices, thus the AlN material system is of great interest due to having the widest direct gap (~6 eV) among all III–V nitrides.[6] There have been very few reports on near-band-edge (NBE) emission (cathodoluminescence) of AlN (Refs. 7 and 8) due to the lack of high-quality materials usually contaminated with oxygen[7] and difficulties associated with this far UV region. In the present work, we report results on a photoluminescence (PL) study of bulk single crystals, as well as of epilayers of wurtzite (WR)-type AlN in a wide range of temperatures (10–300 K), wavelengths (195–600 nm) and excitation intensities (up to a power density of 3 $MW/cm^2$). At room temperature, NBE emission and deep-level PL bands were observed from bulk crystals as well as from 0.3-$\mu$m-thick epilayers (over basal plane sapphire). The analysis of PL measured from *C*-plane (0001) and *A*-plane $(2\bar{1}\bar{1}0)$ together with x-ray measurements clearly showed a higher quality of *A*-plane oriented bulk AlN crystals.

All the bulk SC AlN samples for this study were obtained from Crystal IS. First, AlN boules were grown by a solid-state vapor transport. These were subsequently cut and polished in different orientations yielding the *A*- and the *C*-plane substrates for our study. The AlN epilayer for this study was deposited on a basal plane sapphire substrate using low-pressure MOCVD and trimethyl aluminum (TMA) and $NH_3$ as the precursors. A low-temperature (650 °C) AlN buffer of 100 Å was first deposited followed by the AlN films at 1070 °C with a thickness around 0.3 $\mu$m using a small V/III ratio.[9]

The PL spectra were measured using pulsed excimer laser excitation ($\lambda = 193$ nm, $\tau = 8$ ns, $f_r = 100$ Hz). The laser beam was nearly perpendicular to the *A*- or the *C*-plane surface of the crystal, and the PL signal was collected in a backscattered geometry. The laser beam was focused to a spot of about 0.3 mm diam. The laser light power density could be changed by a set of UV neutral filters. The samples were mounted onto the cold finger of a closed-cycle He cryostat. The PL signals were analyzed using a SPEX 550 monochromator with a UV-enhanced charge-coupled-device array.

Figure 1 shows the room-temperature PL spectra of the studied AlN single crystals for the *A*-plane $(2\bar{1}\bar{1}0)$ and the *C*-plane (0001) surfaces [see inset Fig. 1(a)] and the 0.3-$\mu$m-thick epilayers in a spectral range from 3 to 6.2 eV. As can be seen, it was possible to resolve several groups of PL bands. The short wave emission close to ~6 eV, we believe, is related to the transitions at the fundamental absorption edge, whereas the bands in the region from 3.7 to 4.6 eV are related to the radiative recombination involving deep levels. It was surprising to note that the epilayers demonstrated a weaker deep-level emission intensity in comparison with the bulk material (AlN). As speculated in the past,[7,8] the impu-

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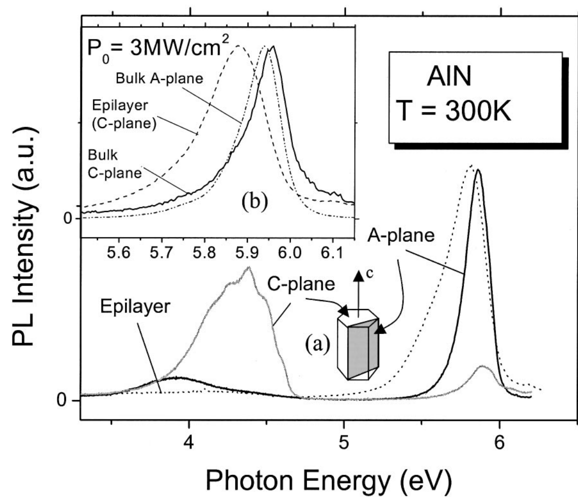


FIG. 1. PL spectra of SC-AlN for *A* and *C* planes and AlN epilayers at room temperature under excitation power density ~1 MW/cm$^2$. Inset (a) shows WR-type crystal *c* axes, *C*-plane (0001), and *A*-plane $(2\bar{1}\bar{1}0)$. Inset (b) shows room-temperature NBE emission spectra of various AlN samples under very high excitation.

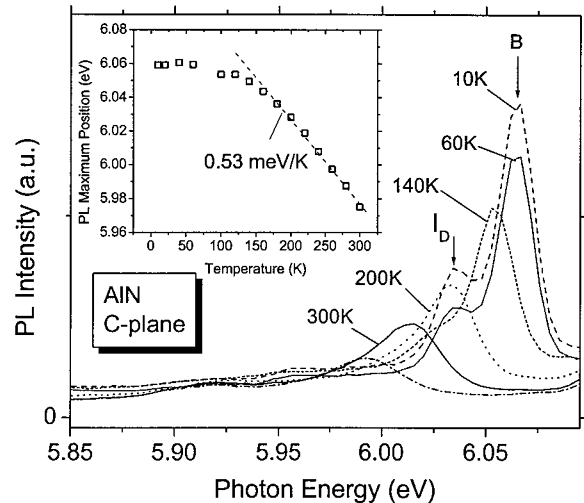


FIG. 3. SC-AlN *C*-plane spectra at different temperatures. *B* denotes the free-exciton line, whereas $I_0$ marks bound exciton line. Inset shows the exciton line position dependence on temperature.

rity related PL band can be assigned to oxygen related defects in AlN. For comparison, in Fig. 1, we also show the *C*-plane bulk-AlN PL that had a predominate deep-level band. Note for this case, similar to the past observations for GaN,[10,11] it was possible to resolve a vibronic structure indicating a strong electron–phonon coupling that might be assigned to optical transitions between donors and acceptors. However, more work is needed for a full identification of deep-level-related optical transitions in AlN.

The fundamental optical transitions near the band edge, in general, dominated the emission spectrum. At low excitations, the various AlN samples of this study had a structure to the NBE emission spectrum. Figure 2 shows an example of such a spectrum for a *C*-plane bulk AlN crystal at room temperature and under different excimer laser excitation intensities. For the excitation power densities below ~500 kW/cm$^2$ we could clearly resolve at least two emission lines located at 5.985 and 5.886 eV. We attribute these lines to the free-exciton radiative annihilation (*B* line) and its LO phonon replica (*B*-LO). The line separation of 99 meV precisely matches the LO phonon energy in AlN.[12] For increased excitation levels the PL-spectrum structure details gradually vanished and a broad band was formed (we denote it as the *P* band). This maximum wavelength position for this band varied with excitation intensity, the type of AlN sample and the *c*-axes configurations. Figure 1(b), inset, shows normalized room-temperature PL of NBE for bulk *A*- and *C*-plane AlN, as well as for the 0.3-$\mu$m-thick AlN epilayer on the sapphire under excitation of ~3 MW/cm$^2$. In the latter case the strongest redshift was observed (the *P*-band maximum is located at 5.88 eV). The *P* bands for *A*- and *C*-plane bulk AlN under the same excitation conditions were located at 5.94 and 5.95 eV, respectively. We believe that the PL for AlN under these strong excitations is due to a radiative recombination from the electron–hole (e–h) plasma. Indeed, e–h plasma band should appear when wave functions of excitons overlap in a dense e–h system and the bound states vanish. Due to bandgap renormalization,[13] the e–h plasma emission band is usually located on the long wave side of exciton lines and it is rather broad. This is similar to the *P*-band behavior for other direct-gap crystals.[14,15] Since the full width at half maximum (FWHM) of the e–h plasma band and its position are determined by the carrier concentration, the e–h plasma model might explain the variety of *P* bands for our different samples under the same excitation conditions. The highest concentration (consequently, the strongest redshift and highest FWHM) is expected in AlN epilayers since carriers are trapped in a layer in contrast to the bulk-AlN crystals where the photogenerated carriers can rapidly penetrate into the bulk. The slight difference of the *P*-band shape and position for the *A*- and *C*-plane bulk AlN [see the spectra in the inset (b) of Fig. 1(b)] can be the result of smaller diffusion lengths for the parallel (*A*-plane), as compared to the perpendicular (*C*-plane), direction with respect to the *c* axis of the crystal.

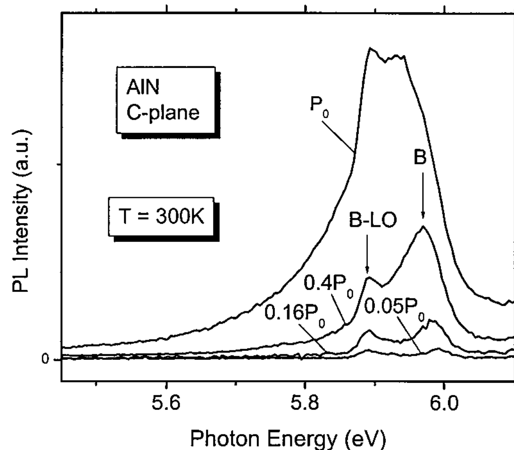


FIG. 2. SC-AlN *C*-plane PL spectra as a function of excitation power density. $P_0 = 2$ MW/cm$^2$.

Figure 3 shows the PL for the *C*-plane bulk AlN as a function of temperature. This behavior is typical for the samples of this study. As can be seen, when the temperature is decreased the PL intensity increases and its maximum position undergoes a blueshift. Simultaneously, a new structure starts to form when the temperature reaches ~150 K. At *T*

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$=10$ K at least two clearly resolved lines can be seen. The short wave free-exciton line at 6.065 eV remains the most intense. The temperature dependence of its position is shown separately in the inset to Fig. 3. The exciton line peak wavelength shift with temperature agrees well with the band-gap change (with temperature) within the interval from 300 to 160 K. It shows an almost linear increase with a slope coefficient of 0.53 meV/K. This is very close to the value obtained cathodoluminescence spectra in Ref. 8. The other low-temperature line ($I_0$), which is located at 6.034 eV at 10 K, can be due to a bound exciton with a rather large (~30 meV) binding energy. Note that some structure of the spectrum (presumably due to LO-phonon replicas) remains on the long wavelength wing of the NBE emission. More experiments are underway for a detailed study of the nature of these exciton complexes in AlN.

In conclusion, the PL of *A*- and *C*-plane bulk crystals and thin epilayers of wurtzite-type AlN was investigated under strong-pulsed excimer-laser excitation. The spectra consisted of a long wave band probably from the deep-level transitions and an intense NBE emission. The NBE emissions were from processes involving free excitons and their LO-phonon replicas at high temperatures and the bound excitons at low temperatures. Under strong excitations a new *P* band forms in the NBE region and is explained by a dense electron–hole plasma model. The strong NBE emission involving exciton-related transitions clearly confirms the high quality of the AlN bulk crystals and the epilayers for UV optoelectronic and microelectronic devices applications.

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