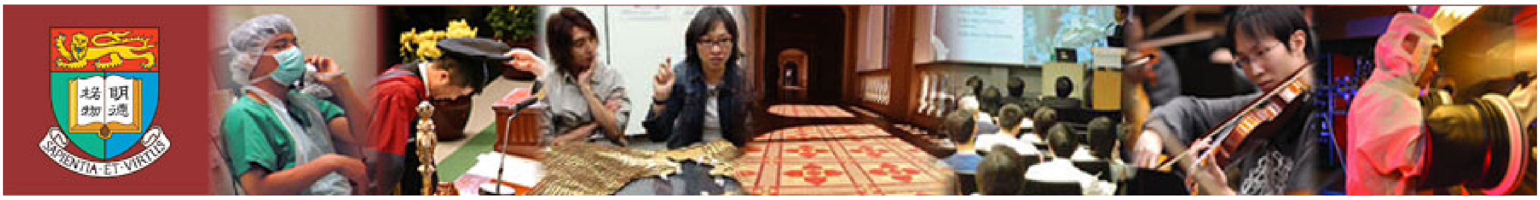

Title	**Shallow optically active structural defect in wurtzite GaN epilayers grown on stepped 4H-SiC substrates**
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Shallow optically active structural defect in wurtzite GaN epilayers grown on stepped 4H-SiC substrates

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A number of wurtzite GaN epilayers directly grown on 4H-SiC (0001) misoriented by 0, 3.5°, 5°, 8°, and 21° with plasma-assisted molecular-beam epitaxy were optically characterized with photoluminescence and excitation spectra. An intense shallow-defect emission peak locating at energy position ~70 meV lower than the near band edge emission peak at 3.47 eV is found in the emission spectra of the GaN films on 4H-SiC misoriented by 8° and 21°. Stacking mismatch boundaries are supposed to be the candidate causing the optical transition. Combined with the low-temperature photoluminescence excitation spectra of the films, the location of the electronic level induced by the structural defect is determined to be about 100 meV above the valence-band maximum of GaN. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1623006]

GaN has been recognized as the leading candidate for the fabrication of shorter wavelength light-emitting devices. Unfortunately, the heteroepitaxy growth of GaN results in the formation of extended defects with density typically as high as $10^{10}\ \mathrm{cm}^{-2}$ in GaN epilayers predominantly due to the lack of lattice-matched substrates.[1,2] Available transmission electron microscopy observation has revealed that threading dislocations, stacking faults (SFs), inversion domain boundaries (IDBs) and stacking mismatch boundaries (SMBs) (also called double positioning boundaries) are several major types of extended defects in GaN epilayers grown on different substrates by means of different growth techniques.[1–4] It is generally believed that these extended defects degrade characteristics of the GaN-based devices and thus their influence on electrical and optical properties of GaN films is certainly required to be understood. In particular, it is essential to know whether they give rise to electrically active defect states in the band gap. Theoretical investigations of the SFs and the IDBs in wurtzite GaN have been reported.[5–9] However, there have been no strong experiments to verify or check such theoretical predictions so far, particularly due to rather complicated situation in the actual GaN heteroepilayers, such as interactions between the different extended defects as well as between the extended defects and impurities in the films.[10,11] Here, a series of GaN thin films are directly grown on vicinal 4H-SiC surfaces with different step densities under same growth conditions with plasma-assisted molecular beam epitaxy. Photoluminescence (PL) and excitation (PLE) optical spectroscopy are then employed to investigate the extended defects in the deposited films. A strong emission peak related to a structural defect is observed in the GaN films grown on 4H-SiC (0001) surface misoriented towards $(11\bar{2}0)$ by 8° and 21°. Its energetic position is determined to be ~104 meV above the valence-band maximum of the GaN.

The substrates used in growth are *n*-type doped 4H-SiC from Nippon Steel Co. Before being loaded into the growth vacuum chamber, they are degreased repeatedly in acetone and ethanol under ultrasonic vibration. Prior to the GaN deposition, the substrates are thermally deoxidized at about 1100 °C in a Si flux, which leads to atomically smooth and clean surfaces. GaN epitaxy will then start by simultaneously supplying the Ga and N source fluxes under the Ga-rich condition. The Ga/N flux ratio is kept at about 2.0. The substrate temperature is set at 650 °C. Under these conditions, GaN is basically grown at a step-flow growth mode.[12] Following 0.5–1.8 μm film deposition, the growth is stopped and the sample is quenched for *in situ* scanning tunneling microscopy observation at room temperature. The PL spectroscopy setup employed in the present work has been described elsewhere.[13] For the PLE measurements, two Acton SP300 monochromators were used as emission and excitation monochromators, respectively. A Müller xenon lamp was used as excitation light source while a Hamamatsu R928 photomultiplier tube was used to detect the emission signal. The samples were mounted on the cold finger of a Janis closed cycle cryostat for measurement of variable-temperature PLE spectra. In the PL and PLE measurements, standard lock-in amplification technique was used to enhance the signal-to-noise ratio. Figure 1 shows the semilogarithmic PL spectra of the GaN films grown on 4H-SiC (0001) surfaces with different miscut angles. The most striking feature in Fig. 1 is that an intense peak at ~3.40 eV (corresponding wavelength ~365 nm) can be observed for the GaN films grown on the 4H-SiC surfaces with the miscut angles of 8° and 21°. For the sample grown on 21° misoriented 4H-SiC surface, the peak is even stronger than the near-band edge transition at ~3.47 eV. But for other samples, such a peak cannot be observed. In order to confirm that it is really from a structural defect in the GaN epilayers, two additional GaN epilayers were grown on the 8° and 21° misoriented 4H-SiC

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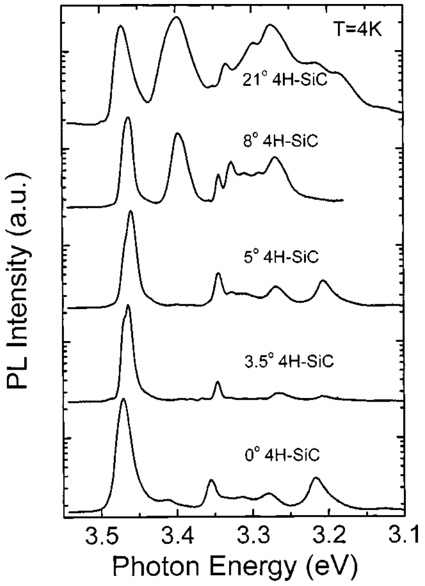

FIG. 1. 4 K PL spectra of the GaN films grown on 4H-SiC (0001) surfaces with different miscut angles.

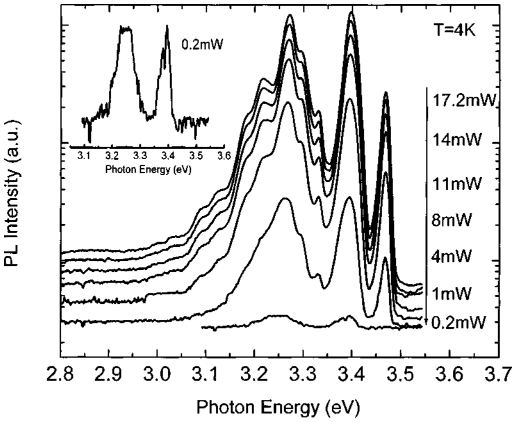

FIG. 2. Excitation-power-dependent PL spectra of the GaN films grown on 4H-SiC (0001) surface with miscut angle of 21°.

surfaces under same growth conditions as those of the previous ones, respectively. As expected, the emission peak at $\sim$3.40 eV can be reproducibly observed in the regrown GaN epilayers. In the present work, we concentrate on discussing the peaks at $\sim$3.40 and 3.27 eV. The 3.27 eV (corresponding wavelength $\sim$379 nm) peak is associated with the zinc-blende phase GaN inclusion in wurtzite GaN host.[14] Notice that both 3.27 and 3.40 eV peaks simultaneously become stronger with increasing the substrate miscut angle, which implies a relationship between the mechanisms behind these peaks. The 3.21 eV peak is argued to be another structural defect related transition in wurtzite GaN.[10] Grandjean *et al.*'s[10] result strongly suggests that the appearance of the 3.21 and 3.40 peaks is very likely due to the structural defects and moreover they are associated with the remaining zinc-blende GaN regions. From Fig. 1, it can be judged that the optical quality of GaN epilayers grown on SiC substrates with the miscut angle range of 3.5°–5° is relatively higher. This conclusion is consistent with the transmission electron microscopy results[12] and is in agreement with other groups' reports.[2,15] These results show that the particulars of atomic steps on substrates strongly affect the structural and optical properties of GaN epilayers.

As mentioned earlier, the GaN films studied in the present work were directly grown on 4H-SiC substrates, which means that there are no buffer layers. In the initial growth stage of GaN, small islands nucleate on the terraces. The coalescence of islands may generate various structural defects. Interestingly, these initial islands may have different atomic stacking sequence because they originate from steps at different levels on the substrate surface. Therefore, the stacking faults and the stacking mismatch boundaries are closely associated with the steps. Recent structural characterization[4] shows that the SF and SMB defects in GaN epilayers really originate from the steps on sapphire and SiC surfaces. In our case, the surfaces of 4H-SiC substrates with large mistcut angles consist of much higher density of steps than the surfaces of substrates with small mistcut angles. It is reasonable and straightforward that higher densities of steps lead to generation of higher density of SF and SMB defects. We thus attribute the appearance of intense 3.40 eV peak and strong increase of the 3.27 eV peak in intensity to an increase of the SF and SMB defect densities in the GaN films deposited on 4H-SiC substrates with large mistcut angles.

The excitation-power and temperature dependence of PL spectra can provide useful information of the emission peaks and hence help to identify their nature. The 4 K PL spectra of the GaN layer grown on 4H-SiC (0001) with 21° miscut angle are measured at different excitation powers, as depicted in Fig. 2. Inset in Fig. 2 is close up of the PL spectrum of the sample under excitation power of 0.2 mW. Like the near-band edge transition (the 3.47 eV peak), the energetic position of the 3.40 eV peak is not excitation-power dependent. In contrast with the 3.47 and 3.40 eV peaks, the 3.27 eV peak seems to exhibit an observable blue-shift with increasing excitation power. For example, the central energetic position of the peak is 3.25 eV when the excitation power is as low as 0.2 mW. It should be noted that there is actually multiple peak structure in this spectral region. It is difficult to identify the origin of each peak in the spectral region. The dominant peak at 3.27 eV is most likely to be the excitonic transition related to the zinc-blende GaN inclusions. Figure 3 shows the PL spectra measured at different temperatures from the GaN film grown on 4H-SiC (0001) with 21° miscut angle. As expected, the near-band edge transition exhibits usual temperature dependence. It is clear that all of the defect-related peaks decrease in intensity faster than the near-band edge peak as the temperature is increased. When the temperature is elevated to 260 K and above, these defect peaks are no longer observable. However, the quenching temperatures of the various defect peaks are different. According to the observed excitation power and temperature dependence of PL spectra shown in Figs. 2 and 3, the 3.40 eV peak should be a transition like free-to-bound (acceptor) transition. The SMBs are the most probable candidate inducing such an acceptor-like level. According to a spatially resolved PL investigation on intentionally grown GaN IDBs, IDBs produce a bright emission peak with energy 30–40 meV below the band edge transition of GaN.[16] The intense new peak observed in our case locates 70 meV lower than the band edge transition. Therefore, the IDBs should be ruled out as candidate causing the 3.40 eV peak. In order to deter-

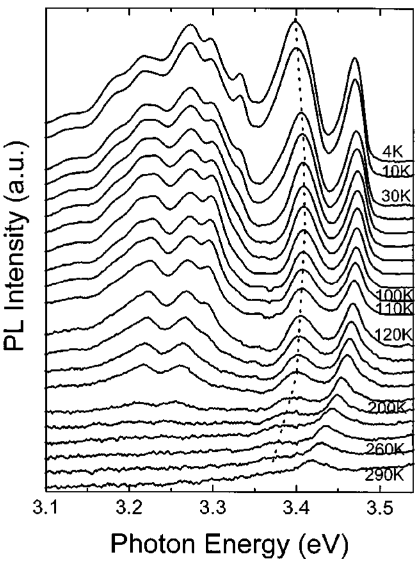

FIG. 3. Temperature-dependent PL spectra of the GaN films grown on 4H-SiC (0001) surface with miscut angle of 21°.

mine the position of electronic level induced by the SMBs, low-temperature PLE spectra of the sample were measured, as shown in Fig. 4. The 10 K PL spectrum is also shown in the figure. It should be noted that the PL spectrum were measured from the sample under excitation of the 345 nm common light dispersed from the xenon lamp. A clear resonance peak at 3.493 eV is observed in the all PLE spectra in spite of different detection energy (marked in wavelength in the figure), which gives the band gap of the GaN epilayer studied in the present work. The energy spacing between the bandgap and the 3.40 eV peak is determined to be ~100 meV. Also, the binding energy of the donor bound excitons producing the band edge transition can be determined to be ~30 meV.

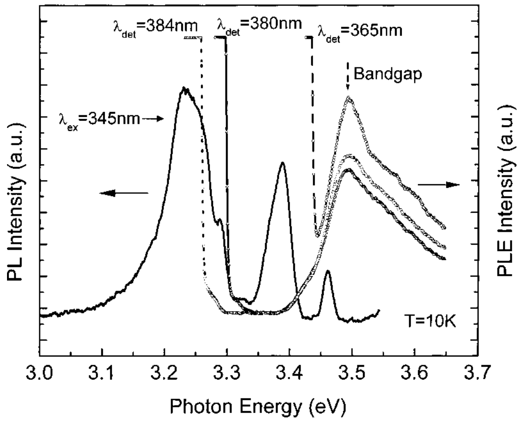

FIG. 4. 10 K PL and PLE spectra of the GaN films grown on 4H-SiC (0001) surface with miscut angle of 21°.

In summary, a series of wurtzite GaN films directly grown on stepped 4H-SiC surfaces were optically characterized. An intense PL peak at ~3.40 eV is observed in the PL spectra of the GaN films deposited on 4H-SiC surfaces with high density of atomic steps. The stacking mismatch boundaries are proposed to be the candidate causing this free-to-acceptor like optical transition. The localized states induced by the structural defect located about 100 meV above the maximum valence band of GaN.

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