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LETTER TO THE EDITOR

Low-pressure chemical vapour deposition growth of high-quality ZnO films on epi-GaN/α-Al_2O_3

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Abstract
We present the first results on (0001) ZnO/(0001) epi-GaN/(0001) α-Al_2O_3 heterostructure fabrication combining metal–organic vapour phase epitaxy and low-pressure chemical vapour deposition methods. The surface morphologies of the films were studied, and x-ray and reflection high-energy electron diffraction measurements were made, which showed a high degree of structural perfection of the ZnO films, with crystallite misorientation as low as $21'$. The measured photoluminescence spectra of the films featured prevailingly emission within the excitonic region.

Wide-band-gap gallium nitride has potential applications in (i) highly efficient light-emitting devices, including blue lasers and devices emitting short-wavelength visible light, and (ii) low-cost solar cells. Another material analogous to GaN is zinc oxide, which is also considered a promising material for applications involving the emission of near-UV and blue light. The large excitonic binding energy (about 60 meV) of ZnO may make it useful in efficient lasing devices based on stimulated emission due to the recombination of excitons at room temperature [1]. For both materials, the main technological problem is that of how to produce defect-free high-quality single-crystalline heterostructures on suitable substrates. As a rule, low-cost sapphire α-Al_2O_3 substrates have been used for such purposes, although there is a significant lattice mismatch between α-Al_2O_3 ($a = 4.754$ Å, $c = 12.99$ Å), GaN ($a = 3.189$ Å, $c = 5.185$ Å), and ZnO ($a = 3.250$ Å, $c = 5.213$ Å). For example, it was reported in [2] that difficulties arose with the application of the CVD technique to basis-oriented ZnO growth onto (0001) α-Al_2O_3 because the crystal lattice mismatch reached as high as 38%. The use of substrates with lower lattice mismatch would improve the quality of the ZnO layers especially near the interface region, and the nearness in lattice parameters of GaN and ZnO allows one to achieve heteroepitaxial growth of good-quality ZnO on GaN and GaN on ZnO. One may also notice a small thermal mismatch because of the slight difference in thermal expansion coefficient between the two materials (6.51×10^{-6} K^{-1} for ZnO and 5.59×10^{-6} K^{-1} for GaN). Another

advantage of this heterostructure would be that the band line-up could be changed by controlling the interface bonds between ZnO and GaN. Although ZnO/GaN heterostructures have such potential advantages, there have been only a few reports on the growth of ZnO on GaN [3, 4] and there are no reports at all on the low-pressure CVD growth of this heterostructure.

We report here on the epitaxial growth of high-quality (0001) ZnO layers on a (0001) GaN-buffered (0001) α-Al_2O_3 substrate in a low-pressure CVD system, with the ZnO/GaN lattice mismatch as low as 1.8%. The degree of structural perfection and the photoluminescence of these heterostructures were also studied.

High-quality epitaxial (0001) GaN buffer layers up to 2 μm thick were grown on (0001) sapphire substrates by the metal–organic vapour phase epitaxy (MOVPE) method using a low-temperature buffer layer technique [5]. These structures, without preliminary polishing and cleaning, were used as templates for ZnO epitaxy in a low-pressure CVD reactor. Hydrogen was used as the working gas in the CVD process. The ZnO films were grown at a substrate temperature ranging from 580 °C to 640 °C, a hydrogen pressure of 10–30 Torr, and with a hydrogen flow of 1 $\ell\ h^{-1}$. The reactor design as well as the temperature optimization route are described elsewhere [6]. ZnO layers 3–5 μm thick were fabricated during the CVD growth.

The surface morphology of the (0001) ZnO epilayer at ×500 magnification displays the regular hexahedrons which are characteristic of the c-plane. This surface pattern is typical for a relatively low substrate temperature, below 600 °C. With temperature increase, the growth patterns become smaller, and they are completely absent at temperatures of 620 °C and above. The film surface becomes mirror-like in this case.

X-ray studies were performed with the DRON diffractometer utilizing Cu Kα radiation. Figure 1 shows the x-ray pattern of 4 μm thick ZnO layer grown onto a GaN/α-Al_2O_3 heterostructure at 625 °C. There are only (0001) reflections of ZnO and GaN in the diffractogram; the reflections of other orientations are absent. It should be noted that the (0002) peaks of ZnO and GaN are not resolved due to a very good lattice matching. It should also be noted that a fine

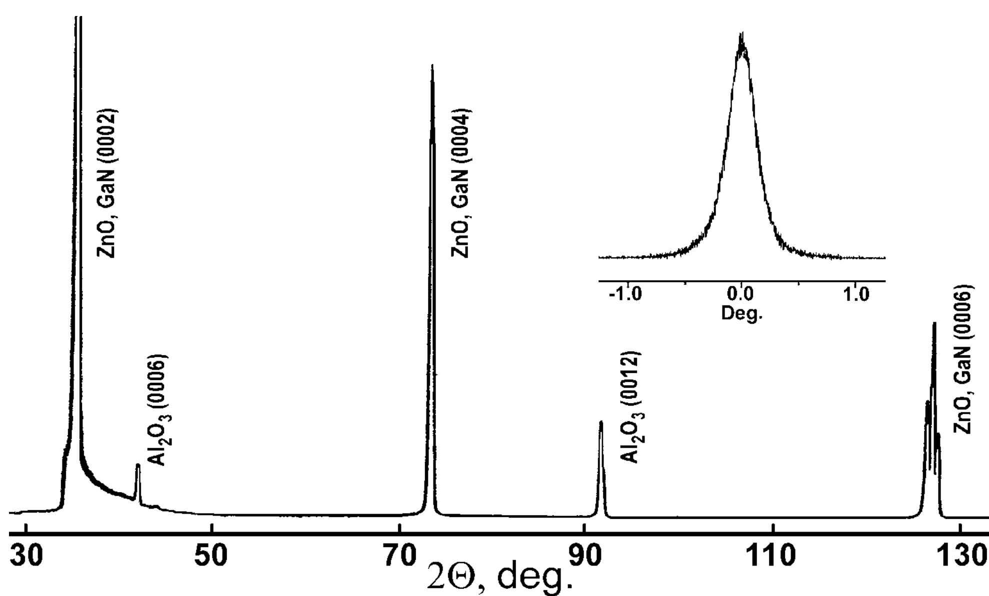

Figure 1. An x-ray pattern of a (0001) ZnO/(0001) epi-GaN/(0001) α-Al_2O_3 heterostructure. The inset shows the rocking curve of the (0002) peak.

structure of the Cu Kα line can be seen in the (0006) ZnO and GaN reflections. The inset in figure 1 shows rocking curve patterns for the (0002) ZnO peak. The full width at half-maximum (FWHM) of the diffraction peak did not exceed $21'$, indicating good ordering along the growth direction. The fact that only the (0001) reflection family can be seen in the diffractogram is not sufficient evidence on which to base a final conclusion of single crystallinity of the film, because with the focusing technique used, only diffraction from the planes parallel to the substrate surface were detected, so additional reflected high-energy electron diffraction (RHEED) measurements have been carried out. Figure 2 shows a RHEED pattern of a 4 μm thick (0001) ZnO layer deposited onto (0001) epi-GaN/(0001) sapphire at 625 °C. The spotty pattern, representing only reflections from the (0001) plane, along with the x-ray diffraction data, conclusively confirmed the high structural quality of the ZnO film. We believe that this is due to the matching of the stacking order and a very low crystal lattice mismatch between ZnO and GaN as compared to the case for layers grown directly on sapphire.

Figure 2. A RHEED pattern for ZnO on (0001) epi-GaN/(0001) α-Al_2O_3.

It was also of interest to study the ultra-violet photoluminescence (PL) of the ZnO layers, because the analysis of the fine structure of the exciton spectra gives additional information on the degree of crystal lattice perfection [7]. Figure 3 shows the characteristic PL spectrum of a film recorded at 77 K using an SPM2 monochromator and a synchronous detection technique. The samples on which measurements were performed were placed directly into liquid nitrogen. It should be pointed out that the UV region in the spectrum showed much higher (more than 30 times) intensity than the green one (inset in figure 3). Such a spectrum relates to a film with perfect structure, in which light is emitted primarily at the A1 free-exciton line and partly within the band of bound excitons J_D (similar spectra were identified in [8]).

In conclusion, use of the (0001) epi-GaN/(0001) α-sapphire heterostructure allows us to fabricate high-quality basis-oriented zinc oxide films by the CVD process in a low-pressure system. The films exhibit perfect structural and good luminescence properties which may be useful for efficient room temperature optically pumped UV laser applications. We ascribe the substantial enhancement of the ZnO film quality first and foremost to the very low lattice mismatch in the ZnO/GaN/α-Al_2O_3 heterostructure.

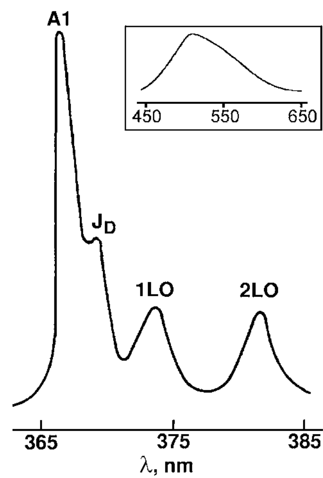

Figure 3. A photoluminescence spectrum taken for basis-oriented 4 μm thick ZnO films deposited onto (0001) epi-GaN/(0001) α-Al_2O_3 at 625 °C.

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