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LETTER TO THE EDITOR

Sharp variations in the temperature dependence of optical reflectivity from AlN/GaN heterostructures

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Abstract
Sharp variations in optical reflectivity were observed when cooling and heating AlN/GaN heterostructures on sapphire substrates between room temperature and 10 K. The reflectivity was found to decrease at a definite temperature T_k in the downward temperature run, and to recover at $T_\mathrm{r} > T_\mathrm{k}$ in the subsequent upward temperature run. The temperature behaviour of reflectivity exhibits memory on the cooling–heating cycles previously subjected to samples.

GaN and related alloys are presently the most attractive materials for fabrication of optoelectronic devices in the ultraviolet and blue energy regions [1, 2]. Apart from this, the AlGaN/GaN heterostructure field-effect transistors (FETs) have emerged as attractive candidates for high-voltage, high-power operation at micro-wave frequencies [3–5]. Due to the lack of suitable nitride substrate material, heteroepitaxial growth on sapphire or 6H-SiC substrates is a common practice. The lattice mismatch between GaN and substrate materials causes the growth of GaN layers exhibiting a specific morphological feature, namely mosaicity or domain structure [6]. From crystallographic point of view, the submicrometre-size single crystalline domains are usually tilted and/or rotated (twisted) with respect to each other [6, 7]. According to [8], a GaN epilayer may be considered as a heterogeneous system consisting of three phases, namely of single crystalline GaN grains, inter-granulated material and pipes (voids). The morphology of GaN epilayer, in its turn, strongly influences the crystalline quality of the top AlN film.

In this work, we used optical reflectivity to characterize AlN/GaN heterostructures in the temperature range 10–300 K. Sharp temperature-induced changes in optical reflectivity were observed which are believed to be initiated by the variation of strains with temperature.

The GaN and AlN layers were grown by low-pressure (60–110 Torr) metalorganic chemical-vapour deposition (MOCVD) on *c*-plane (0001) sapphire substrates. Standard precursors of trimethylgallium (TMGa), trimethylaluminium (TMAl) and ammonia (NH_3) were used as alkyl and hydride sources [9]. The alkyl and hydride sources were kept separate until reaching the quartz reactor. The carrier gas was Pd-cell purified hydrogen (H_2). Heating was accomplished by RF induction of the graphite susceptor. Switching of all valves and manifolds was done using computer control. The sapphire substrates were initially cleaned in trichloroethylene, acetone, isopropyl alcohol and H_2SO_4/H_3PO_4. After a high-temperature (1200 °C) cleaning in H_2, the growth temperature was lowered to 500 °C. Nitridation was performed and then an $\sim$20 nm thick GaN nucleation layer was grown. After ramping the temperature to 1100 °C, a 1300 nm unintentionally doped GaN channel layer was grown. The thickness of the top AlN film was 5 nm.

The quality of the GaN layer and the presence of the AlN thin film were confirmed using a high-resolution x-ray diffractometer (HRXRD). The $\theta/2\theta$ XRD scans have shown

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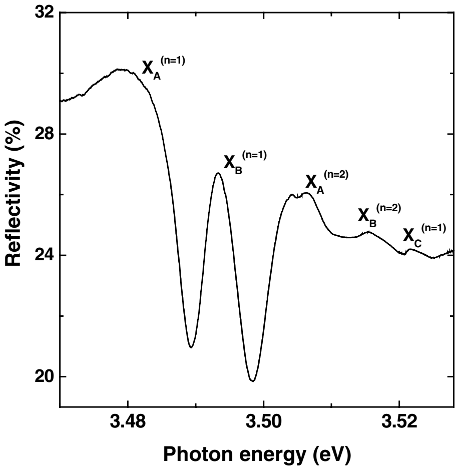

Figure 1. Reflectivity spectrum of GaN in the exciton region $T = 10$ K.

distinct (0002) GaN and AlN peaks, confirming the presence of the AlN layer. The full width at half maximum (FWHM) of the GaN (0002) $\theta/2\theta$ peak was ~120 arcsec. In addition, the (0002) and (10–12) rocking curves for the GaN layer were measured. The FWHM of these peaks equals 372 and 470 arcsec respectively. These values correlate with analogous literature data [10] which is indicative of good quality of the GaN channel layer.

The reflectivity was measured using light from a 100 W halogen lamp. The reflected white light from the sample was analysed through a double spectrometer with 1200 lines/mm gratings equipped with a photomultiplier. Well resolved interference oscillations were evidenced in the transparency spectral range of our GaN layers. To avoid contribution from interference effects, the temperature dependence of reflectivity was studied at a wavelength of 350 nm, i.e. at quantum energy higher than the band gap of GaN. The specimens were mounted on the cold station of LTS-22-C-330 workhorse-type optical cryogenic system.

Figure 1 presents a typical reflectivity spectrum of GaN layers grown as described above. The spectrum consists of two major features at 3.484 and 3.494 eV related to the $X_A^{n=1}$ and $X_B^{n=1}$ free exciton ground states as well as of weak features at higher energies related to $X_A^{n=2}$ and $X_B^{n=2}$ exciton excited states and to the ground state of the $X_C^{n=1}$ exciton. The energy position of the major exciton features in the reflectivity spectrum of our GaN layers is shifted by about 8 meV towards high energies in comparison with that inherent to GaN epilayers grown on GaN substrates [11] or to thick unstressed heteroepitaxial gallium nitride layers [12]. It means that our GaN layers grown on sapphire suffer from biaxial compressive strain. Using the previously reported 20 meV GPa^{-1} rate of the exciton line shift with the biaxial stress in GaN layers [13], one can estimate the value of strains in our layers. It equals 0.4 GPa at 10 K.

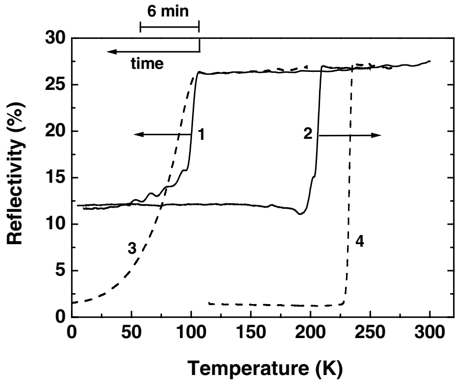

Figure 2. Temperature dependence of reflectivity from AlN/GaN structure measured in downward (curve 1) and upward (curve 2) temperature runs. Curve 3 represents the relaxation of reflectivity with time after the cooling was stopped at 105 K, and curve 4 is the temperature dependence of reflectivity during subsequent heating up to 300 K. The rate of cooling and heating was 6 K min^{-1}.

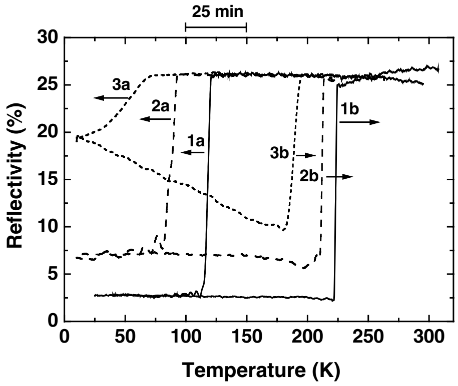

Figure 3. Temperature dependence of reflectivity from AlN/GaN structure during three alternating cooling–heating cycles. The rate of cooling and heating was 2 K min^{-1}.

Figure 2 shows the temperature dependence of reflectivity measured from an AlN/GaN structure in downward (curve 1) and upward (curve 2) temperature runs at a rate of 6 K min^{-1}. Upon decreasing the temperature, the reflectivity exhibits a sharp decrease at $T_k = 105$ K and after some damped oscillations it reaches a new level which is more than twice lower than that observed at 300 K. In the subsequent upward temperature run, the reflectivity recovers at $T_r = 205$ K. The critical temperature T_k does not depend upon the rate of the temperature decrease, but the low-reflectivity state is influenced by this parameter. The reflectivity at low temperatures changes monotonously from 2 to 14% when the cooling rate increases from 0.2 to 10 K min^{-1}. One should note that the transition from the high-reflectivity to the low-reflectivity state in decreasing temperature is initiated by the temperature decrease up to the critical value T_k. After reaching the critical temperature, it occurs independently either that the temperature continues to decrease or it is stopped at $T_k = 105$ K. In the last case (illustrated by curve 3) the reflectivity decreases with time, and it reaches a value of 1.5% in approximately 12 min. As to the recovery of reflectivity, it occurs in the temperature interval 205–230 K, the value of T_r depending upon the level of reflectivity achieved on temperature decrease (figure 2).

Another feature of the phenomenon involved is the sample memory on previous temperature cycles, see figure 3. After the first temperature cycle (curve 1a in decreasing temperature and curve 1b in increasing temperature) the hysteresis loop is shifted to lower temperatures in the second (curves 2a and 2b) and third (curves 3a and 3b) temperature cycles. At the same

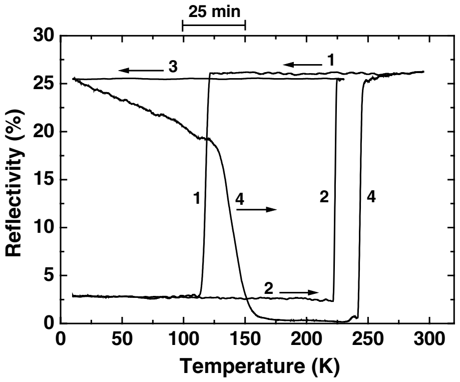

Figure 4. Temperature dependence of reflectivity from AlN/GaN structure measured in two alternating cooling–heating cycles. During the first cycle the temperature increase was stopped at 230 K. The rate of cooling and heating was 2 K min^{-1}.

time, the low-temperature reflectivity is higher for the second respectively third cycles. The memory effect is observed provided that the time between two adjacent temperature cycles does not exceed 20 h. The higher the rate of temperature decrease or increase, the more pronounced the effect. For instance, the 20% level of the low-temperature reflectivity can be achieved in two instead of three temperature cycles by increasing the rate of cooling or heating from 2 to 6 K min^{-1}.

A manifestation of the sample memory on the previous temperature cycles is the possibility to freeze the high-reflectivity state of the sample down to low temperatures as illustrated in figure 4. If after the first cooling (curve 1) the temperature is increased up to a value a little higher than T_r (curve 2) and, after that, the sample is cooled again (curve 3), the level of reflectivity inherent to high temperatures is frozen. However, this state is metastable at low temperatures, and the reflectivity will relax extremely slowly with time (in about 20 h) to a low-reflectivity state. On the other hand, if the temperature is increased before any relaxation starts, the sample can be transferred to a state exhibiting very low reflectivity (curve 4). In such a case, the initial high-reflectivity state recovers upon heating up to 240 K.

The decrease in reflectivity at low temperatures is accompanied by an increase in the efficiency of diffuse light scattering in samples observed by a naked eye. A possible explanation of the phenomenon involved is the following.

Upon decreasing the temperature, the strain in the GaN epilayer increases due to the difference in thermal expansion coefficients of GaN and sapphire. Taking into account the mosaicity of GaN epilayers grown on sapphire substrates, one can suggest that the contrast in reflective indices inside and at the boundary of the domains increases with decreasing temperature. This may happen due to strain-induced transformations of the lattice in high-defective regions between the single crystalline GaN grains. No essential changes seem to occur in the single crystalline grains at the point of transition, since the same set of excitons was found to be inherent to both low- and high-reflectivity states of GaN.

In conclusion, sharp variations in optical reflectivity were observed when cooling and heating the AlN/GaN heterostructures between room temperature and 10 K. The reflectivity was found to decrease at a definite temperature T_k in the downward temperature run, and to recover at $T_r > T_k$ in the subsequent upward temperature run. One can summarize the following regularities related to the phenomena involved: (i) the processes are reversible; (ii) the reflectivity exhibits memory on previous cooling–heating cycles; (iii) the lower the value of reflectivity at low temperatures, the higher the temperature of the reverse transition. The observed phenomena seem to be caused by the domain structure of GaN epilayers grown on sapphire substrates, namely by strain-induced intensification of the contrast in the refractive indices inside and at the boundary of the domains. To get a better understanding of the temperature-induced spectacular changes in properties of AlN/GaN heterostructures, further studies are needed.

Acknowledgments

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