

## Raman studies of hexagonal GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N multilayered structures

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One of the most efficient, sensitive, and direct techniques which can be used for the quantitative characterization of low-dimensional semiconductor structures is Raman spectroscopy. Nevertheless, there are few works devoted to studies of the phonon behavior in multilayered structures based on III-nitrides. The goal of our work was to reveal specific features in the behavior of optical phonons in hexagonal multilayered GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N structures grown by the MOCVD and MBE techniques.

The objects were three multilayered structures grown on sapphire substrates. Two MOCVD-grown samples consisted of 37 pairs of GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers grown on a thin Al<sub>x</sub>Ga<sub>1-x</sub>N buffer layer. The Al content in the first sample was  $x=0.19$ , and the thicknesses of GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers were 39 nm and 45 nm, respectively. The Al content in the second sample was  $x=0.29$ , and the GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers were 37 nm and 53 nm thick, respectively. The sample grown by plasma-assisted molecular beam epitaxy (PAMBE) in a Riber 32 MBE system consisted of 10 pairs of GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N layers with  $x=0.44$ . The thicknesses of the GaN and the Al<sub>x</sub>Ga<sub>1-x</sub>N layers in this sample were 37 nm and 44 nm, respectively. All structures were characterized by X-ray diffraction (XRD) and electron probe microanalysis (EPMA). Raman spectra of the samples were measured in a backscattering configuration at room temperature. An Ar<sup>+</sup> laser ( $\lambda=488$  nm) was used as a source of excitation.

There are six optical phonon modes A<sub>1</sub>(TO), A<sub>1</sub>(LO), E<sub>1</sub>(TO), E<sub>1</sub>(LO), E<sub>2</sub>(high), and E<sub>2</sub>(low) active in the first-order Raman scattering from hexagonal GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N. The phonon energies of Al<sub>x</sub>Ga<sub>1-x</sub>N exhibit a one-mode type behavior in the range of Al concentrations studied and considerably differ from the phonon energies in GaN. Therefore, it could be expected that the Raman spectrum would have two six-line sets if the optical phonons are “confined” to GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers.

In the experiment we observed two E<sub>2</sub>(low) phonons with frequencies corresponding to GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N constituting the multilayered structure. We also observed Al<sub>x</sub>Ga<sub>1-x</sub>N phonons of A<sub>1</sub>(LO) and E<sub>1</sub>(LO) symmetry and the GaN-like LO phonon occupying the intermediate position between the energies corresponding to A<sub>1</sub>(LO) and E<sub>1</sub>(LO) phonons in bulk GaN. However, the A<sub>1</sub>(TO), E<sub>1</sub>(TO), and E<sub>2</sub>(high) phonons were detected only as single lines occupying the intermediate positions between the frequencies of corresponding phonons in bulk GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers.

Our results indicate that propagating phonons rather than confined A<sub>1</sub>(TO), E<sub>1</sub>(TO) and E<sub>2</sub>(high) modes were observed in Raman scattering from multilayered structures. This finding is consistent with the theoretical predictions [1] that optical phonons in a GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N low-dimensional structures can be regarded as propagating due to the energy overlap between optical phonon regions of GaN and the alloy. Long wavelengths of the phonons participating in the Raman scattering suggest that the lattice dynamics parameters averaged over the multilayered structure period should be used to describe the behavior of phonons in the GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N structure. The averaging procedure can be performed by using one of the variants of the isodisplacement approach, which describes well the phonon modes at the  $\Gamma$ -point for many solid solutions. To check the validity of this approach, we compared our experimental data with the data obtained for bulk Al<sub>x</sub>Ga<sub>1-x</sub>N with the Al content equal to the averaged Al content in the multilayered structure  $\bar{x} = ax/(a+b)$ , where  $a$  and  $b$  are the thicknesses of the Al<sub>x</sub>Ga<sub>1-x</sub>N and GaN layers, respectively. The agreement between experimental data and the data corresponding to the averaged Al content for A<sub>1</sub>(TO), E<sub>1</sub>(TO) and E<sub>2</sub>(high) phonons looks acceptable.

However, our observations of GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N phonons of E<sub>2</sub>(low) symmetry, Al<sub>x</sub>Ga<sub>1-x</sub>N phonons of A<sub>1</sub>(LO) and E<sub>1</sub>(LO) symmetry, and GaN-like LO phonon have shown that these optical phonons are “confined” to the GaN or Al<sub>x</sub>Ga<sub>1-x</sub>N layer. Probably this can be caused by an insufficient energy overlap between optical phonon regions of GaN and the alloy. The theoretical approach describing the behavior of all optical phonons in the multilayered structure of this type is in progress.

To summarize, Raman spectroscopic studies of the phonon modes behavior in multilayered GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N structures grown by the MBE and MOCVD techniques have been carried out for the first time. It has been found that the observed A<sub>1</sub>(TO), E<sub>1</sub>(TO), and E<sub>2</sub>(high) phonons can be regarded as propagating over the multilayered structure. At the same time, we have found that E<sub>2</sub>(low) and LO modes are “confined” to GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers. We have demonstrated that Raman spectroscopic data can be used to estimate the Al content in the alloy and the ratio between thicknesses of the GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N layers composing the multilayered structure.

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### References

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