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Optical phonons in hexagonal GaN/Al_xGa_(1-x)N multilayered structures

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Abstract. The results of the phonon behavior in multilayered periodic structures GaN/Al_xGa_(1-x)N are presented. It is shown that the A₁(TO), E₁(TO), and E₂(high) phonons can be considered as the vibrations propagating through the multilayered structure.

Introduction

Recently GaN/Al_xGa_(1-x)N MQW structures have been grown by MBE, and now great efforts are directed towards fabrication of GaN/Al_xGa_(1-x)N superlattices. At present there is a growing need for precise non-destructive characterization of these nanostructures. One of the most efficient, sensitive, and direct techniques which can be used for the quantitative characterization of semiconductors is Raman spectroscopy. Nevertheless, there are few works devoted to studies of the phonon behavior in low-dimensional GaN/Al_xGa_(1-x)N structures. Theoretical treatment of phonons in such structures is restricted to consideration of superlattices (SL) consisting of several monolayers. However, it can be expected that in thicker structures with a period of tens nanometers the phonon behavior will also have specific features due hexagonal symmetry of GaN and AlN and crossing of their optical bands. The goal of our work was to reveal the peculiarities in the behavior of optical phonons in hexagonal multilayered GaN/Al_xGa_(1-x)N structures grown by the MOCVD and MBE techniques.

1. Samples and experimental procedure

The objects were three multilayered structures grown on sapphire substrate. Two structures were grown by MOCVD and one structure was grown by MBE. The MOCVD-grown samples consisted of 37 pairs of GaN and Al_xGa_(1-x)N layers grown on a thin Al_xGa_(1-x)N buffer layer. The Al content in the first sample was $x = 0.24$, and the thicknesses of GaN and Al_xGa_(1-x)N layers were 31.3 nm and 46.2 nm, respectively. The Al content in the second sample was $x = 0.29$, and the GaN and Al_xGa_(1-x)N layers were 37.4 nm and 53.1 nm thick, respectively. The details of the growth can be found in Ref. [1]. The third sample consisting of 10 pairs of Al_{0.53}Ga_{0.47}N/GaN layers was grown by plasma-assisted molecular beam epitaxy (PAMBE) in a Riber 32 MBE system. The thicknesses of the GaN and the Al_{0.53}Ga_{0.47}N layers were 36.9 nm and 43.9 nm, respectively. All three structures were characterized by X-ray diffraction (XRD) and electron probe microanalysis (EPMA). Raman spectra of the multilayered structures were measured in a backscattering

configuration at room temperature. An Ar^+ laser ($\lambda = 488 \text{ nm}$) was used as a source of excitation.

2. Experimental results and discussion

There are six optical modes $1A_1(\text{TO}) + 1A_1(\text{LO}) + 1E_1(\text{TO}) + 1E_1(\text{LO}) + E_2(\text{low}) + E_2(\text{high})$ active in the first-order Raman scattering in hexagonal GaN and $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$. The Γ -point phonon frequencies are well studied for both compounds [2–4]. The results described below refer to phonons of the $A_1(\text{TO})$, $E_1(\text{TO})$ and $E_2(\text{high})$ symmetry which have the highest intensity in the Raman spectrum, and therefore their behavior can be traced in detail.

Figure 1 shows Raman spectra of the multilayered structures for the scattering geometry corresponding to the $A_1(\text{TO})$ phonon. In addition, Raman spectra of bulk GaN and $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ layers grown on a sapphire substrate are given. Note that the Al content in bulk $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ layers was the same as in the structures studied. One can see that in the multilayered structures the $A_1(\text{TO})$ phonon is detected only as a single line occupying an intermediate position between the frequencies of the $A_1(\text{TO})$ phonons in bulk GaN and $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ layers. We have also found that the situation is the same for the $E_1(\text{TO})$ and $E_2(\text{high})$ phonons in multilayered structures.

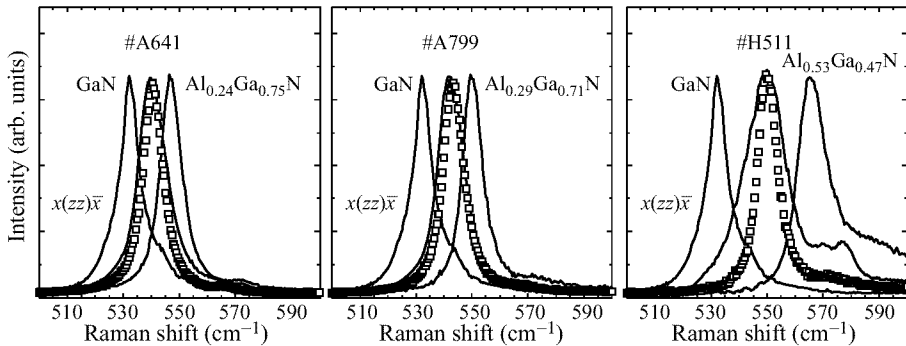


Fig. 1. Raman spectra in the scattering configuration corresponding $A_1(\text{TO})$ phonon mode for different samples.

To our knowledge, only one paper concerned with Raman studies of hexagonal GaN/AlN SL has been published [5]. The authors assigned the observed $E_1(\text{TO})$ and $E_2(\text{high})$ lines to the phonons of GaN layer and explained the blue shift of these lines with respect to their positions for bulk GaN by the built-in strain. However, the position of the line corresponding to the $A_1(\text{TO})$ phonon was not consistent with calculations which included only the SL deformation. As an alternative, the interface origin of this line was suggested in this paper. The absence of lines corresponding to phonons in an AlN layer was attributed to a considerable difference in the Raman cross sections.

In our case the Raman cross sections for the layers comprising the multilayered structures were comparable, and two lines should be observed for a confined phonon of GaN and alloy. Nevertheless, the $A_1(\text{TO})$, $E_1(\text{TO})$, and $E_2(\text{high})$ phonons were observed as single lines, which contradicts the hypothesis of phonon confinement. According to theoretical predictions [6], optical phonons in the AlN/GaN low dimensional structure can be considered as propagating due to the energy overlap between optical phonon regions of GaN and AlN. Our results indicate that propagating phonons rather than confined modes were

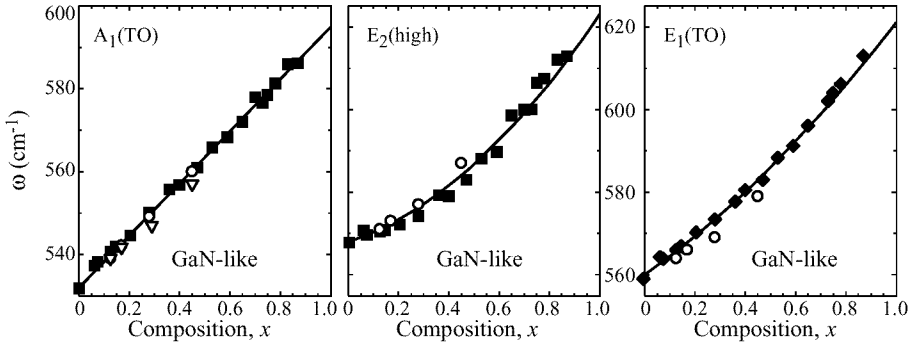


Fig. 2. The behavior of the optical phonon modes as a function of Al content.

observed in Raman scattering from multilayered $\text{GaN}/\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ structures.

Large wavelengths of the phonons participating in the Raman scattering suggest that the lattice dynamics characteristics averaged over the multilayered structure period should be used to describe phonons in the $\text{GaN}/\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ structure. The averaging procedure can be performed by using one of the variants of the isodisplacement approach, which for many solid solutions describes well the phonon modes at the Γ -point. To check the validity of this approach, we compared our experimental data and the data obtained for bulk $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ 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 $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ and GaN layers, respectively. Positions of the experimental maxima for $A_1(\text{TO})$, $E_2(\text{high})$ and $E_1(\text{TO})$ phonons in $\text{GaN}/\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ structures at corresponding \bar{x} (shown by open circles), and experimental compositional dependence of these phonons for bulk $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ are plotted in Fig. 2. Additionally, Fig. 2 presents the positions of the phonon frequencies corresponding to the averaged Al content for the AlN/GaN structure studied in [6]. The agreement between experimental data and the data corresponding to the averaged Al content looks acceptable. The propagating phonons are supposed to be insensitive to the strain due to the different signs of strains in GaN and $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ layers.

We have also estimated the $A_1(\text{TO})$ phonon position in our multilayered structures using the approach developed in [7, 8]. For the $A_1(\text{TO})$ phonon in the $\text{GaN}/\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ structure this approach gives the dielectric function in the form

$$\left[\left\langle \frac{1}{\varepsilon(\omega)} \right\rangle \right]^{-1} = \frac{\varepsilon_a(\omega)\varepsilon_b(\omega)(a + b)}{a\varepsilon_b + b\varepsilon_a}, \quad (1)$$

where ε_a and ε_b are the low-frequency dielectric functions of $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ and GaN , respectively. This approach gives two TO-modes; the low-frequency TO-mode is presented in Fig. 2 (triangle). Its position is seen to coincide well with the position of the corresponding mode of bulk $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ with the Al content equal to that averaged over the structure \bar{x} .

3. Conclusion

To summarize, Raman spectroscopic studies of phonon modes behavior in multilayered $\text{GaN}/\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ structures grown by the MBE and MOCVD techniques have been carried out for the first time. It has been found that the observed the $A_1(\text{TO})$, $E_1(\text{TO})$, and $E_2(\text{high})$ phonons can be considered as propagating over the multilayer structure. We interpret the $A_1(\text{TO})$ mode in the multilayered structure as the vibration modified by interaction

between phonon modes in GaN and $\text{Al}_x\text{Ga}_{(1-x)}\text{N}$ in this structure through a macroscopic electric field.

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