

Micro-Raman-Scattering Experiments of GaN Layers Deposited on Sapphire and SiC

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We show the ability of micro-Raman-spectroscopy to provide information about local orientation, strain and doping levels of GaN layers on the μm scale. Our micro-Raman measurements reveal that a portion of the GaN layer near the substrate interface is oriented in such a way that the c-axis of the GaN layer lies in the interface plane. We have performed micro-Raman measurements in order to test if the free-carrier concentration varies spatially for samples which are semiisolating determined by integral methods. We found spatial differences in the doping levels by two orders of magnitude.

1. Introduction

A main problem in growing GaN layers is the huge lattice mismatch between GaN and usual substrates as e.g. sapphire and 6H-SiC [1]. This causes a drastic disturbance of the crystal quality of the GaN layer which has a lot of defects located directly at the substrate interface as well as penetrating through the whole layer. Consequently, the GaN layers exhibits strong inhomogeneities. The aim of this paper is to show the ability of micro-Raman-scattering experiments to provide information about local orientation, strain and free-carrier concentrations of GaN layers in the μm range. Knowing the free-carrier concentrations may, for example, be helpful when making precise statements about the optical and electrical properties. First, we present results from our spatially-resolved measurements about structural properties and then about doping inhomogeneities.

2. Experimental

The samples used in this work were grown on sapphire by hydride vapor phase epitaxy (HVPE) and on SiC by a modified sandwich technique [2]. All samples possessed wurtzite structure and were undoped except for one Fe-doped layer. Their thickness vary between 50 μm for the samples grown on SiC and 220 μm to 400 μm for the layers deposited on sapphire.

The spatially-resolved micro-Raman experiments were carried out using a Dilor XY800 triple-grating spectrometer and a Dilor LABRAM single-grating spectrometer working with a super-notch filter. Both systems have a charge-coupled device (CCD) as detectors and are equipped with confocal optics. The sample was excited either parallel (in-plane) or perpendicular (on-plane) to the substrate surface using the 488 nm line of an Ar⁺-Kr⁺ mixed-gas laser or the 632.8 nm line of a He-Ne laser. The micro optics focused the laser on a point spot of about 1 μm and detected the scattered light in backscattering geometry, which corresponds to an $x(\cdot)\underline{x}$ configuration for in-plane excitation and a $z(\cdot)\underline{z}$ configuration for on-plane excitation. The samples were cooled down to 4.2 K using an Oxford

microscope cryostat. With this setup we obtained a spatial resolution of about $1\ \mu\text{m}$ and a spectral resolution better than $1\ \text{cm}^{-1}$ for the triple-grating and about $2\ \text{cm}^{-1}$ for the single-grating spectrometer.

Infrared reflectivity measurements at room temperature were performed using a Bruker 66V Fourier transform infrared (FTIR) spectrometer.

3. Results

In Fig. 1 we show a linescan across the GaN/sapphire interface where we have taken a spectrum every $1\ \mu\text{m}$. The sample was cooled down to $4.2\ \text{K}$ and in-plane excited which corresponds to a scattering geometry $x(\cdot)x$. The region of the sapphire substrate is marked by the appearance of the A_g sapphire mode at $419\ \text{cm}^{-1}$. The transition to the GaN layer is indicated by the appearance of the $A_1(\text{TO})$ and the E_2 modes at $534\ \text{cm}^{-1}$ and $569\ \text{cm}^{-1}$, respectively. Directly at the interface to the substrate as well as in a distance about $30\ \mu\text{m}$ away from the interface two photoluminescence bands both peaking at $2.4\ \text{eV}$ appear. The photoluminescence bands are seen as constant in Raman shift background, since they are broad compared to the spectral range observed here. The photoluminescence properties of this sample are described in more detail in [3].

Of great importance is the spatial change in scattering intensity of the different phonon modes. Near the interface to the substrate both the $A_1(\text{TO})$ mode and the E_2 mode are visible; in this region the $A_1(\text{TO})$ is the strongest mode. The ratio of their intensity inverts with increasing distance from the substrate interface. While near the substrate interface the spectra are dominated by the $A_1(\text{TO})$ mode the intensity of the E_2 mode increases with the appearance of the second photoluminescence band and becomes the most intense phonon mode. For clarity three spectra taken at several distances to the substrate interface are shown in Fig. 1.

Considering the selection rules in wurtzite crystals [4] the inversion of the relative scattering intensities can be explained by a structural reorientation of the GaN layer. Near the substrate interface

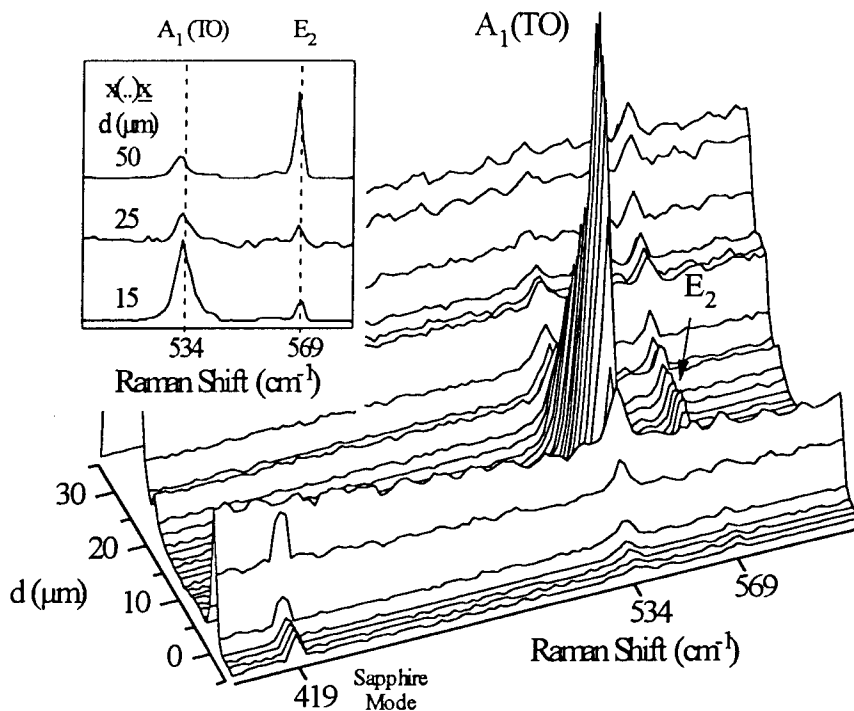


Fig. 1 Linescan across the GaN/sapphire interface of a $400\ \mu\text{m}$ thick sample taken at $4.2\ \text{K}$. The inset shows three Raman spectra taken at different distances to the interface. The inversion of the scattering intensities of the two Raman modes can clearly be seen.

the GaN layer is oriented in such a way that the c -axis of the GaN layer lies in the interface plane. In some μm away from the substrate interface the layer reorients to the *ordinary* growth direction in which the c -axis of the layer is parallel to the growth direction.

We could confirm this structural behavior by on-plane Raman measurements in which we excited the sample parallel to the growth direction, i. e. the k vector of the exciting light is perpendicular to the substrate interface plane (scattering geometry $z(\cdot)z$). We performed these measurements with the 632.8 nm line of a HeNe laser on different GaN samples grown on sapphire as well as on SiC. The confocal optics allows to record Raman spectra at several depths, i. e. distances to the substrate interface. Such a depth profile is shown in Fig. 2. The spectra are dominated by the E_2 and $A_1(\text{LO})$ modes which are the modes predicted by the selection rules for the $z(\cdot)z$ scattering geometry used in this experiment. Near the substrate interface also the $A_1(\text{TO})$ mode becomes clearly visible. This confirms that the c -axis of the layer is not parallel to the growth direction. Hence it follows from both in- and on-plane Raman measurements that at least a portion of the GaN layer near the interface to the substrate is oriented parallel to it.

As can be seen in the inset of Fig. 1 both the E_2 and the $A_1(\text{TO})$ mode shifts by about 2cm^{-1} to lower frequencies while the layer reorients. This behaviour can be explained by strain relaxation as described in Ref. [5]. Kozawa et al. measured the relation between the GaN layer thickness and the phonon frequency and observed similar shifting of the E_2 mode.

The well-known correlation between the free-carrier concentration and the plasma-frequency on the one hand and the LO phonon-plasmon (LPP) coupled modes on the other hand allows to determine the free-carrier concentration of semiconductors using infrared reflectivity measurements or Raman spectroscopy. Perlin et al. have recently applied this method on GaN crystals [6]. They find a one-to-one correspondence between the plasma edge obtained from infrared spectra and the high-frequency LPP modes obtained from the Raman experiments.

Infrared as well as the standard transport measurements are integral methods in determining the free-carrier concentration and it is interesting, e.g. for electronic applications, to test if GaN layers exhibit homogeneous doping levels. We show that micro-Raman spectroscopy is an excellent tool to investigate the spatial distribution of the free-carrier concentration in GaN layers.

For this purpose we have first performed infrared reflectivity experiments on our GaN samples in order to obtain an integral value of the free-carrier concentration by measuring the plasma edge. For the GaN layer we want to discuss in the following, which was iron doped in order to lower the free-carrier density by compensation, we obtained by FTIR at room temperature a free-carrier concentration of about $5.1 \cdot 10^{17} \text{cm}^{-3}$. Electronic parametric resonance spectroscopy (EPR) carried out confirmed that

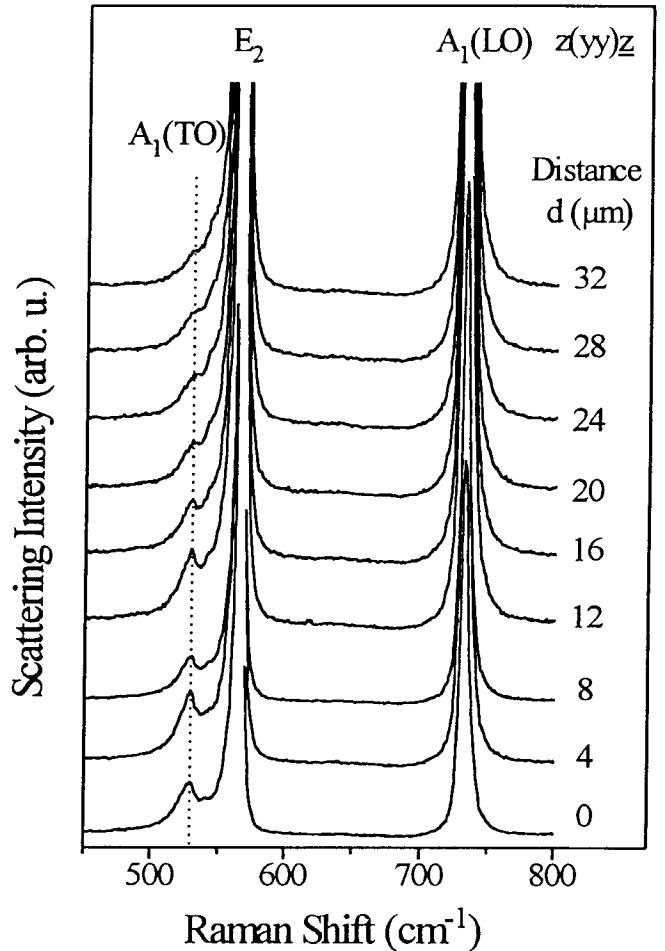


Fig. 2 Depth profile of a 220 μm thick GaN layer grown on sapphire. The Raman spectra are taken at room temperature using the 632.8 nm line of a He-Ne laser.

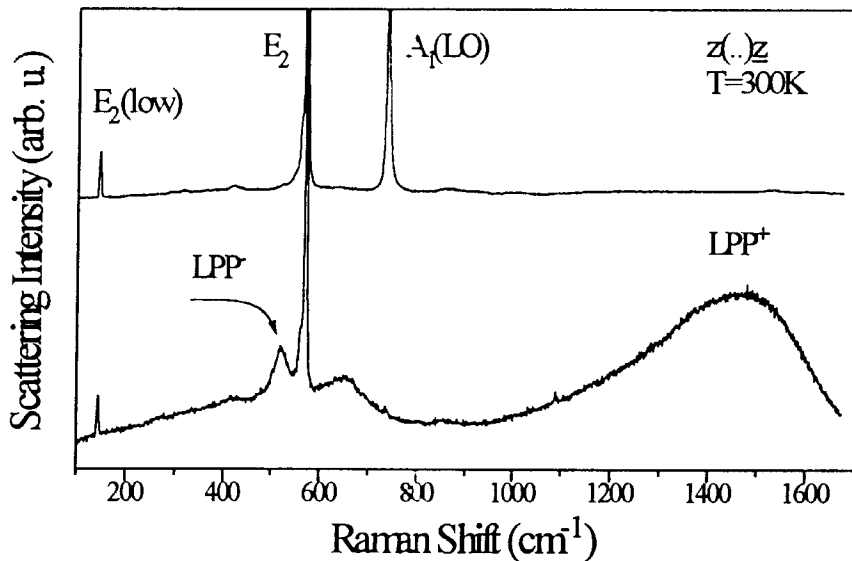


Fig. 3: Room-temperature Raman spectra taken at different points on a GaN/sapphire sample.

the free-carrier concentration is in the semiinsulating range. In Fig. 3 we show micro-Raman spectra taken at different points of the sample in $z(\cdot)z$ geometry. The biggest part of the sample exhibits Raman signals as shown in the upper spectrum. The allowed E_2 modes and the $A_1(\text{LO})$ mode at 735cm^{-1} are the only modes observed. On the other hand in regions extending only few μm we obtained spectra as shown in the lower curve. The E_2 mode remains the strongest Raman mode in the spectrum but no $A_1(\text{LO})$ mode could be detected. Instead of the LO mode two broader structures located at 520cm^{-1} and 1470cm^{-1} can be seen. They can clearly assigned to the low- and high-frequency LPP mode, respectively [6]. Using the correlation between the LPP frequency and the plasma frequency given in [6] for GaN we obtained a local free-carrier concentration of about $2.5 \cdot 10^{19}\text{cm}^{-3}$. Thus, the free-carrier concentration in this sample differs by two orders of magnitude.

4. Conclusion

We have shown, that micro-Raman spectroscopy is an excellent method in providing information about local changes of the orientation and strain in GaN layers. We found that a portion of the GaN layer near the substrate interface is oriented in such a way that the c -axis of the GaN layer lies in the interface plane. Our measurements allows to determine spatial distribution of the free-carrier concentration in such layers with a spatial resolution of about $1\ \mu\text{m}$. We found differences in the free-carrier concentration by two orders of magnitude.

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