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Effect of anisotropic strain on phonons in *a*-plane and *c*-plane GaN layers

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Abstract

We have studied phonons in two types of anisotropically strained GaN films: *c*-plane GaN films grown on *a*-plane sapphire and *a*-plane GaN films grown on *r*-plane sapphire. The anisotropic strain in the films is determined by high-resolution X-ray diffraction (HRXRD) in different measuring geometries and the phonon parameters have been assessed by generalized infrared spectroscopic ellipsometry (GIRSE). The effect of strain anisotropy on GaN phonon frequencies is presented and the phonon deformation potentials $a_{A_1(\text{TO})}$, $b_{A_1(\text{TO})}$, $c_{E_1(\text{TO})}$ and $c_{E_1(\text{LO})}$ are determined.

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1. Introduction

During the last three decades group-III nitrides have continuously attracted strong research interest due to their unique properties and application in optoelectronics, high-power and high-frequency electronics. Despite the remarkable progress in the field, growth of group-III nitrides still suffers from the lack of native substrate and nitride-based technology vastly relies on heteroepitaxy. The growth on foreign substrates typically leads to the presence of built-in strain in heteroepitaxial nitride layers due to the difference in lattice parameters and thermal expansion coefficients between layers and substrates. Sapphire and SiC are among the most often used substrates and typically growth is realized on the basal (0001) *c*-plane of sapphire and SiC. In such instances nitride films grow along the [0001]

direction and experience isotropic strain. Whenever growth is realized on non-*c*-plane sapphire and SiC the nitride films will be under anisotropic strain (independent of the film orientation) as a consequence of the anisotropy of the hexagonal (nitrides, SiC) and rhombohedral (sapphire) crystal structures.

Strain has a significant impact on fundamental properties and device-relevant characteristics of materials. The effect of anisotropic strain on the optical response and electronic band structure has been experimentally studied for *c*-plane GaN [1,2], *a*-plane GaN [3,4] and *m*-plane GaN films [5,6]. The impact of anisotropic strain on lattice parameters of *c*-plane GaN films grown on *a*-plane sapphire [7] and on the structural properties of *a*-plane GaN [8,9] has been also reported. In contrast, the vibrational properties of anisotropically strained GaN are poorly studied and no effect of anisotropic strain on phonons has been reported. Theory predicts that some of the group-III nitride phonons split under anisotropic strain

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[10], since the GaN $[1\ 1\ \bar{2}\ 0]$ and $[1\ \bar{1}\ 0\ 0]$ directions are no longer equivalent in terms of strain as it is in the isotropic case. However, no experimental evidence has been given. Furthermore, no information on the c phonon deformation potentials, related to the strain anisotropy in the basal hexagonal plane exists. In addition, non- c -axis oriented GaN allows an access to the complete set of polar phonons by infrared spectroscopic ellipsometry (IRSE), which provides an alternative or complementary tool to Raman spectroscopy when studying vibrational properties. In this respect it is worth mentioning that there is a lack of agreement between some phonon deformation potentials of GaN and AlN as determined by theory [10] and by employing Raman scattering spectroscopy (RS) [11,12], which makes further clarification of these issues by independent techniques highly desirable.

The goal of this work is to study the effect of anisotropic strain on the phonon modes of GaN films exhibiting different strain patterns and to determine the phonon deformation potentials. The phonon parameters have been assessed by generalized IRSE (GIRSE), and the anisotropic strain in the films is determined by high-resolution X-ray diffraction (HRXRD) in different measuring geometries.

2. Strain determination

We studied two types of anisotropically strained GaN layers: (i) c -plane GaN films grown on a -plane sapphire by metalorganic chemical vapor phase epitaxy (MOVPE) and hydride vapor phase epitaxy (HVPE) with thicknesses of 2 and 40 μm , respectively. The MOVPE films were grown in an Aixtron planetary reactor at 1170 °C using a low temperature nucleation GaN layer and the HVPE growth is carried out at 1080 °C without using a buffer layer. (ii) a -plane GaN films grown on r -plane sapphire by MOVPE and HVPE. In this case films with different thicknesses between 1 and 67 μm were studied. The a -plane MOVPE films were grown at 1030–1050 °C using a conventional two-step process employing GaN or AlN buffer layers [13]. The a -plane HVPE films were grown directly on sapphire in a three-zone horizontal reactor at 1040–1070 °C [14].

HRXRD was performed using a Philips triple axis diffractometer following the alignment procedure described in Ref. [15]. Both, a and c lattice parameters of the GaN layers were measured. The c lattice parameters of all films were determined from the 0002, 0004 and 0006 $2\theta - \omega$ diffraction peaks. In the case of c -plane GaN films the measurements were performed for four different azimuthal positions achieved by a rotation of the sample about its normal by 90°. For the a -plane GaN films the measurements were performed in edge symmetric geometry at two azimuths. The two a lattice parameters in the case of c -plane GaN films were determined from the asymmetric 10 $\bar{1}$ 4, 10 $\bar{1}$ 5, 20 $\bar{2}$ 4 and 20 $\bar{2}$ 5 reflections at all six azimuth positions and the average value of the c lattice parameter. In the case of a -plane GaN films the a lattice parameter

along the growth direction was determined from the 11 $\bar{2}$ 0 and 22 $\bar{4}$ 0 reflections for four azimuth positions separated by 90°. The a lattice parameter lying in the growth plane was determined from the 1 $\bar{1}$ 00 and 2 $\bar{2}$ 00 peaks measured in edge symmetric geometry. Details about the lattice parameter determination of the c - and a -plane GaN films can be found in Refs. [7,9], respectively.

The GaN films grown on both a - and r -plane sapphire are expected to experience anisotropic strain in the basal plane because of the difference in thermal expansion coefficients between sapphire and GaN along the GaN $[1\ 1\ \bar{2}\ 0]$ and $[\bar{1}\ 1\ 0\ 0]$ directions as well as the different lattice mismatches. Accordingly, we have experimentally determined two different a lattice parameters for our GaN films. In the case of c -plane oriented films we found that the $2\theta - \omega$ positions in the asymmetric scans appear at different positions, when measured at the six azimuthal positions, reflecting a distortion of the hexagonal symmetry [7]. Four of the $2\theta - \omega$ positions appear at lower and two at higher scattering angles. The values of the interplanar distances derived from these peak positions can be discriminated accordingly into two groups and consequently, the in-plane lattice parameter also has two different values [7]. The two a lattice parameters are smaller than the strain-free value [16] as one can expect from the difference in the thermal expansion coefficients between GaN and sapphire [17]. The distortion of the GaN basal plane under this anisotropic compressive strain is illustrated in Fig. 1(a). The c lattice parameters of the c -GaN films was found to be larger than the strain-free value as the elasticity theory predicts in the case of biaxial strain. Similarly to the c -oriented films, we found that all a -plane GaN films are compressively strained in the growth plane (both along the $[0\ 0\ 0\ 1]$ and $[1\ \bar{1}\ 0\ 0]$ directions) and the strain along the growth direction is tensile. The strain in this case however results in more complex distortion of the basal GaN plane, which is compressed along the $[1\ \bar{1}\ 0\ 0]$ and expanded along the $[1\ 1\ \bar{2}\ 0]$ directions as illustrated in Fig. 1(b).

3. Modeling of the GIRSE data. GaN phonons under anisotropic strain

The vibrational properties of all films were studied by room-temperature GIRSE. The GIRSE measurements were performed in the spectral range of 350–1500 cm^{-1} with a spectral resolution of 2 cm^{-1} , and at 60° and 70° angles of incidence. In the case of c -plane GaN films the measurements at each angle of incidence were performed for different angles between the plane of incidence and the GaN $[1\ 1\ \bar{2}\ 0]$ direction. For the a -plane GaN films the GIRSE spectra at each angle of incidence were taken at different angles between the plane of incidence and the c -axes of the films. For our films it is necessary to apply a generalized ellipsometry approach since both GaN and sapphire are anisotropic materials and because of the non- c -axis orientation of the substrates or/and films [15,18].

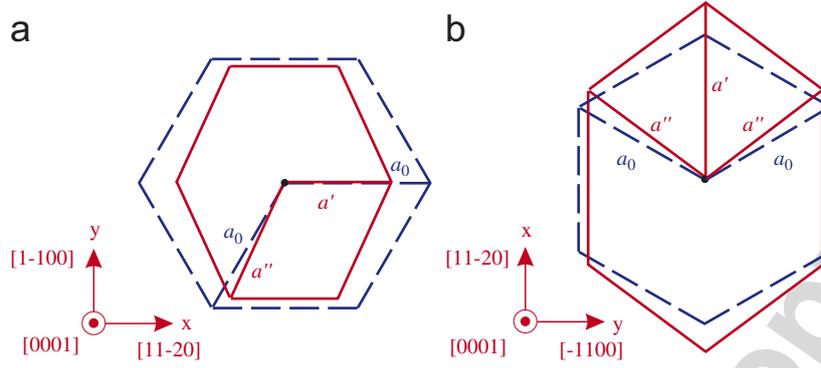


Fig. 1. Schematic drawings of the distortion of the GaN basal plane under anisotropic strain: *c*-plane GaN on *a*-plane sapphire (a) and *a*-plane GaN on *r*-plane sapphire (b).

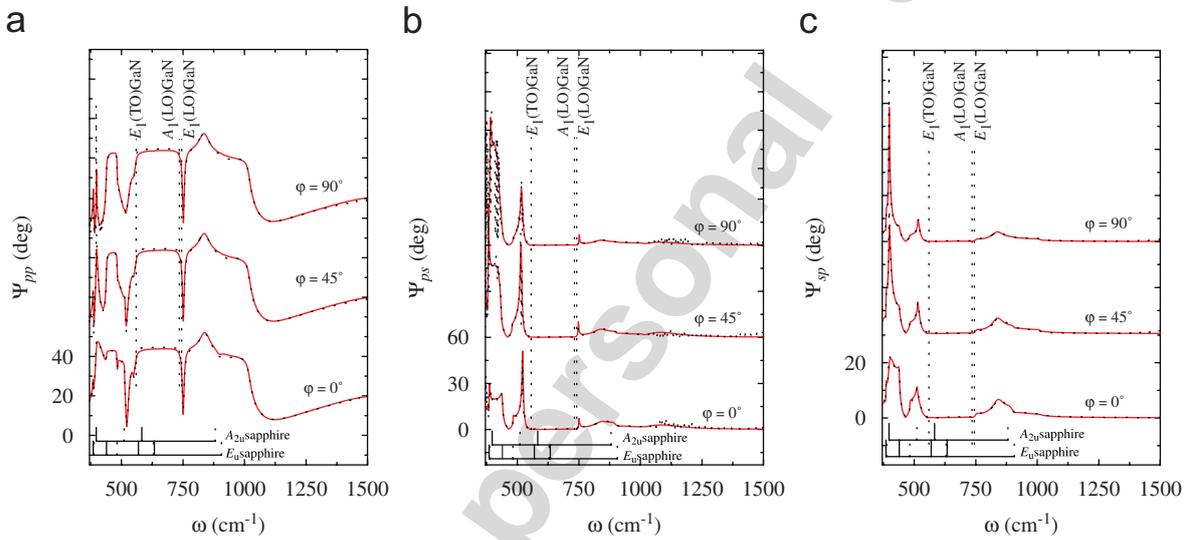


Fig. 2. Experimental (dots) and calculated (solid lines) GIRSE spectra of one representative *c*-plane GaN film on *a*-plane sapphire for different angles ϕ between the plane of incidence and the GaN $[1\ 1\ \bar{2}0]$ direction: (a) Ψ_{pp} ; (b) Ψ_{ps} and (c) Ψ_{sp} . The frequencies of the GaN phonon modes of A_1 and E_1 symmetry, as well as the A_{2u} and E_u phonon modes of sapphire are indicated.

A regression analysis is necessary to obtain the IR optical properties from the GIRSE data. The GIRSE data were modeled using an anisotropic GaN dielectric function parameterized along ($j : z = [0001]$) and perpendicular ($j : x = [1\ 1\ \bar{2}0]$; $j : y = [1\ \bar{1}\ 00]$) to the GaN *c*-axis accounting for the polar phonon contribution

$$\epsilon_j^L(\omega) = \epsilon_{\infty,j} \frac{\omega^2 + i\gamma_{LO,j}\omega - \omega_{LO,j}^2}{\omega^2 + i\gamma_{TO,j}\omega - \omega_{TO,j}^2}, \quad (1)$$

where $\omega_{TO,j=x,y,z}$ denote the frequencies of the $E_1^x(\text{TO})$, $E_1^y(\text{TO})$ and $A_1(\text{TO})$, while $\omega_{LO,j=x,y,z}$ denote the frequencies of the $E_1^x(\text{LO})$, $E_1^y(\text{LO})$ and $A_1(\text{LO})$ modes. $\gamma_{TO,j}$, and $\gamma_{LO,j}$ are the corresponding mode broadening parameters, and $\epsilon_{\infty,x}$, $\epsilon_{\infty,y}$ and $\epsilon_{\infty,z}$ are the high-frequency limits of the dielectric function for polarization perpendicular and parallel to the GaN *c*-axis. A possible contribution from free-carriers to the dielectric function of the GaN layers [19].

Fig. 2 shows experimental and best-fit model calculated Ψ_{ij} spectra for one representative *c*-plane GaN layer on *a*-plane sapphire. The off-diagonal elements Ψ_{ps} and Ψ_{sp} are mostly needed to locate the exact in-plane orientation of the sapphire *c*-axis during the GIRSE measurements. The model parameters that were allowed to vary during the GIRSE data analysis are the frequency and broadening parameters of the $E_1^x(\text{TO})$, $E_1^y(\text{TO})$, $E_1^x(\text{LO})$, $E_1^y(\text{LO})$ and $A_1(\text{LO})$ modes, the high-frequency limit of the dielectric function, $\epsilon_{\infty,j}$, the carrier concentration and mobility, and the thickness of the layers. Equal broadening parameters $\gamma_{TO,j} = \gamma_{LO,j}$ were assumed for $j = x, y$. The IRSE data are not sensitive to the TO resonance frequency with polarization vector parallel to the sample normal. Consequently, the $A_1(\text{TO})$ phonon mode resonance of *c*-plane GaN could not be found and the values determined from RS measurements were used in the calculated data as fixed parameters.

The GIRSE spectra of the *c*-plane GaN films are dominated by the reststrahlen bands of sapphire, indicated

by the brackets below the spectra in Fig. 2. The sharp peaks in Ψ_{ij} spectra within the sapphire reststrahlen range (300–980 cm^{-1}) are due to bands of total reflection for the p and s polarized light components between the E_u and A_{2u} phonon modes. The lattice modes of the GaN films also affect the Ψ_{pp} spectra. In particular, the GIRSE spectra provide high sensitivity to the frequency and broadening parameter of the GaN $E_1(\text{TO})$ mode. It is seen in Fig. 2 that a distinct spectral feature in Ψ_{pp} appear near the $E_1(\text{TO})$ resonance. A spectral feature in Ψ_{pp} related to the LO mode has also been detected for our films and $A_1(\text{LO})$ and $E_1(\text{LO})$ phonon mode parameters can be obtained from the line-shape analysis. The frequencies and the broadening parameters of the phonons polarized in the x - y plane and the high-frequency dielectric constants for polarization along $[1\ 1\ \bar{2}\ 0]$ and $[\bar{1}\ 1\ 0\ 0]$ directions, extracted from the best-fit modeling to the experimental data, are given in Table 1. A subtle difference in the frequency values of the E_1 phonons was determined for polarizations along the GaN $[1\ 1\ \bar{2}\ 0]$ and $[\bar{1}\ 1\ 0\ 0]$ directions upon model layer calculations. This experimental finding agrees with the theoretical predictions that the frequencies of the GaN E_1 phonons are shifted and split with respect to the strain-free values [10]. In the linear strain limit these shifts and splittings are related to the strain tensor via [10]

$$\Delta\omega_{E_1} = a_{E_1}(\varepsilon_{xx} + \varepsilon_{yy}) + b_{E_1}\varepsilon_{zz} \pm c_{E_1}[(\varepsilon_{xx} - \varepsilon_{yy})^2 + 4\varepsilon_{xy}^2]^{1/2}. \quad (2)$$

The coefficients, a_{E_1} , b_{E_1} and c_{E_1} are the corresponding mode deformation potentials per unit strain. Since the x -, y - and z -axes are chosen parallel to the GaN $[1\ 1\ \bar{2}\ 0]$, $[\bar{1}\ 1\ 0\ 0]$ and $[0\ 0\ 0\ 1]$ principal axes, respectively, the shear stress and strain components ($i \neq j$) are zero. Using our experimental results for the strain components and the phonon frequency splittings we have estimated for the first time the c phonon deformation potential for the $E_1(\text{TO})$ and $E_1(\text{LO})$ modes to be 379 and 682 cm^{-1} , respectively.

It can be seen from Table 1 that the broadening parameters of the E_1 phonons polarized along the $[1\ 1\ \bar{2}\ 0]$ and $[\bar{1}\ 1\ 0\ 0]$ directions also show a difference of about 0.6 cm^{-1} . The $E_1^x(\text{TO})$ and $E_1^x(\text{LO})$ exhibit larger broadening which could be related to the larger compressive in-plane strain along the GaN $[1\ 1\ \bar{2}\ 0]$ direction as compared to the $[\bar{1}\ 1\ 0\ 0]$ direction. The high-frequency dielectric

Table 1
Best-fit values of the $E_1(\text{TO})$ and $E_1(\text{LO})$ phonon frequencies and broadening parameters, and high-frequency dielectric constants for polarizations along the GaN $[1\ 1\ \bar{2}\ 0]$ and $[\bar{1}\ 1\ 0\ 0]$ directions

Polarization	ω (cm^{-1})		γ (cm^{-1})	$\varepsilon_{\infty,\perp}$
	$E_1(\text{TO})$	$E_1(\text{LO})$		
$[1\ 1\ \bar{2}\ 0]$	$559.8 \pm .04$	$745.2 \pm .05$	$9.2 \pm .1$	$5.18 \pm .01$
$[\bar{1}\ 1\ 0\ 0]$	$559.3 \pm .04$	$744.3 \pm .04$	$8.6 \pm .07$	$5.15 \pm .01$

constant also shows a subtle anisotropy for the polarizations in the x - y plane, most probably related to strain effects as well.

For the a -plane GaN films the model fitting parameters are the frequencies and broadening parameters of the GaN $E_1^x(\text{TO})$, $A_1(\text{TO})$, $E_1^x(\text{LO})$, $E_1^y(\text{LO})$ and $A_1(\text{LO})$ modes, the high-frequency limit of the dielectric function, $\varepsilon_{\infty,j}$, the carrier concentration and mobility, and the thickness of the layer. In this case the GIRSE data are not sensitive to the GaN $E_1^x(\text{TO})$ resonance and its frequency and broadening parameter were fixed to those of the $E_1^y(\text{TO})$ mode. Experimental and best-fit model calculated Ψ_{ij} spectra for one representative a -plane GaN layer on r -plane sapphire are shown in Fig. 3. The coupling between p- and s-polarized light, sensed by the off-diagonal elements, is small at $\phi = 0^\circ$ and minimal at $\phi = 90^\circ$ (Figs. 3(b) and (c)). A noticeable conversion of p-(s-) to s-(p-) polarized light related to the GaN film is observed in the GIRSE spectra for intermediate ϕ , particularly across frequency regions of resonant excitation of TO phonons or LO-related anisotropic Fano surface polariton and surface guided wave modes. It can be seen that the low-frequency edge of the GaN reststrahlen band in Ψ_{pp} moves from $E_1(\text{TO})$ to $A_1(\text{TO})$ when the azimuth angle, ϕ , between the plane of incidence and the GaN c -axis changes between 0° and 90° (Fig. 3(a)). The $E_1(\text{TO})$ polarized along $[\bar{1}\ 1\ 0\ 0]$ and $A_1(\text{TO})$ are excited resonantly depending on ϕ , as the electric field parallel to the film interface senses lattice excitations with polarization perpendicular or parallel to the GaN c -axis, respectively.

We extracted from the best-fit to the GIRSE data the frequencies and broadening parameters of the polar phonons, as well as the free-carrier concentrations and mobilities for all samples. We focus on the $A_1(\text{TO})$ phonons, as they were so far studied only theoretically or by RS experiments. The $A_1(\text{TO})$ mode frequency obeys the following relation with strain.

$$\Delta\omega_{A_1(\text{TO})} = a_{A_1(\text{TO})}(\varepsilon_{xx} + \varepsilon_{yy}) + b_{A_1(\text{TO})}\varepsilon_{zz}, \quad (3)$$

where $a_{A_1(\text{TO})}$ and $b_{A_1(\text{TO})}$ are the $A_1(\text{TO})$ phonon deformation potentials. A regression analysis of the experimental frequencies and strains is necessary to obtain the deformation potentials and strain-free frequency. However, Eq. (3) cannot be directly used since the three strain components are not independent. As the stress component along the growth direction vanishes because the surface is free to expand or contract it can be shown that

$$\varepsilon_{yy} = -\frac{C_{11}}{C_{12}}\varepsilon_{xx} - \frac{C_{13}}{C_{12}}\varepsilon_{zz} \quad (4)$$

and consequently

$$\Delta\omega_{A_1(\text{TO})} = a_{A_1(\text{TO})} \left(1 - \frac{C_{11}}{C_{12}} \right) \varepsilon_{xx} - \left(a_{A_1(\text{TO})} \frac{C_{13}}{C_{12}} - b_{A_1(\text{TO})} \right) \varepsilon_{zz}. \quad (5)$$

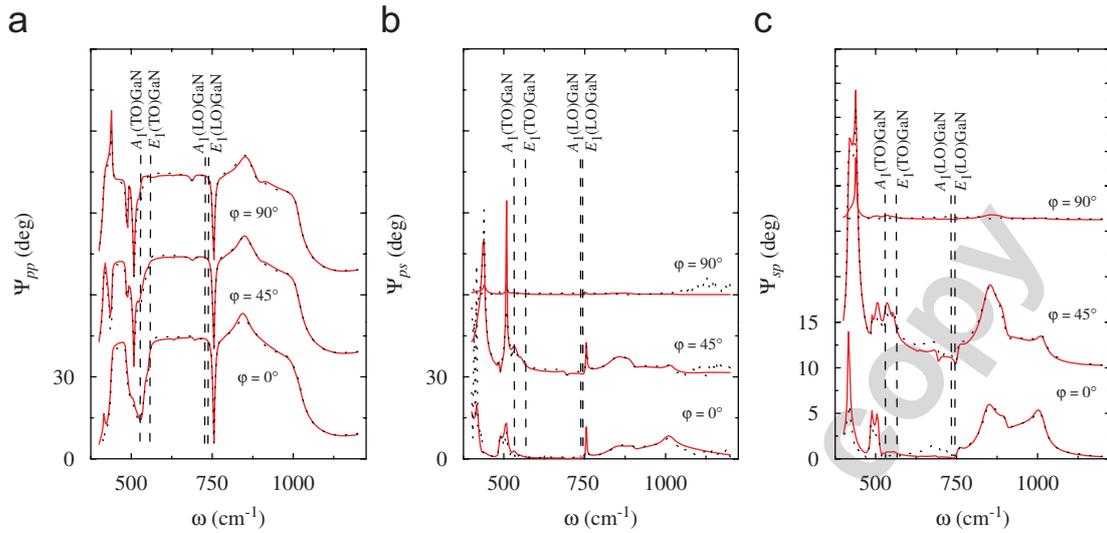


Fig. 3. Experimental (dots) and calculated (solid lines) GIRSE spectra of one representative *a*-plane GaN film on *r*-plane sapphire for different angles ϕ between the plane of incidence and the GaN [0001] direction: (a) Ψ_{pp} ; (b) Ψ_{ps} and (c) Ψ_{sp} . The frequencies of the GaN phonon modes of A_1 and E_1 symmetry are indicated.

Table 2

$A_1(\text{TO})$ strain-free frequency, ω_0 , and deformation potentials, $a_{A_1(\text{TO})}$ and $b_{A_1(\text{TO})}$

References	ω_0 (cm ⁻¹)	$a_{A_1(\text{TO})}$ (cm ⁻¹)	$b_{A_1(\text{TO})}$ (cm ⁻¹)
This work	531.2	-664	-1182
Raman exp. [11]	531.8	-630	-1290
Theory [10]	540	-640	-695

Then using our experimental results about the $A_1(\text{TO})$ frequency, ϵ_{xx} , ϵ_{zz} and the stiffness constants from Ref. [20] we performed a regression analysis of Eq. (5) in order to obtain the phonon deformation potentials and the strain-free frequency of the GaN $A_1(\text{TO})$ mode. The obtained values are listed in Table 2 and compared with previous theoretical and experimental results. It is seen that there is a very good agreement between the strain-free frequency values obtained by GIRSE and RS, while the theoretical value slightly deviates. The $a_{A_1(\text{TO})}$ obtained by GIRSE in the present work is in a good agreement with RS and theoretical results that have been previously reported [10,11]. Furthermore, the $a_{A_1(\text{TO})}$ obtained by GIRSE in the present work is in a good agreement with the RS and theoretical results [10,11]. Our result for $b_{A_1(\text{TO})}$ agrees very well with the value determined previously from RS experiments, confirming the previously reported large difference between theoretical estimations and experiment [10,11]. The reasons for the observed difference are not well understood but we note that a similar discrepancy between theory and experiment was found to be a general tendency for other phonon modes of GaN and AlN as well [10].

4. Conclusions

The effect of anisotropic strain on GaN phonons was studied for *c*-plane GaN on *a*-plane sapphire. It was found that the anisotropic in-plane strain leads to splitting of the GaN $E_1(\text{TO})$ and $E_1(\text{LO})$ phonons. This finding is in agreement with theoretical predictions and presents the first experimental evidence. It also allows us to estimate for the first time the *c* phonon deformation potential of the GaN $E_1(\text{TO})$ and $E_1(\text{LO})$ phonons to be 379 and 682 cm⁻¹, respectively. We have also studied the vibrational properties of *a*-plane GaN layers with different thicknesses in correlation with the anisotropic strain in the films. As a result we determined the GaN $A_1(\text{TO})$ deformation potentials to be $a_{A_1(\text{TO})} = -664$ cm⁻¹ and $b_{A_1(\text{TO})} = -1182$ cm⁻¹, and the strain-free $A_1(\text{TO})$ frequency to be 531.2 cm⁻¹.

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