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Comparative Study of the Electronic Band Structure of Strained *C*-plane and *M*-plane GaN Films by Polarized Photoreflectance Spectroscopy

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We have investigated a strained *C*-plane GaN(0001) film on 6H-SiC(0001) and a strained *M*-plane GaN($1\bar{1}00$) film on γ-LiAlO$_2$(100) by polarized photoreflectance spectroscopy, wherein the electric-field vector **E** of the probe beam is varied in the film plane. In the *C*-plane film, **E** always remains perpendicular to the unique *c* axis of wurtzite GaN, and the spectral features do not change with polarization rotation. In the *M*-plane film, the orientation of **E** changes relative to **c**, and the spectral features are significantly altered. Using the Bir–Pikus Hamiltonian, we calculate the energy and the oscillator strength components of the three transitions at the band gap of GaN as a function of an arbitrary strain in the *C* plane and the *M* plane. On its basis, we can explain the origin and the polarization properties of the spectral features in both films.

Introduction Recently there has been an increased interest in *M*-plane GaN ($1\bar{1}00$) (cf. Fig. 1a). Unlike in *C*-plane GaN(0001), electrostatic fields due to spontaneous and piezoelectric polarization are absent in *M*-plane films. This can lead to more efficient quantum-well-based light emitters [1]. The surface normal to an *M*-plane film is perpendicular ($\perp$) to the unique **c** axis of wurtzite GaN, and optical properties of such films are therefore expected to depend strongly on the in-plane polarization of a normally incident light beam. In addition, strain in the film affects the electronic band structure (EBS) and can significantly alter the polarization selection rules for optical transitions.

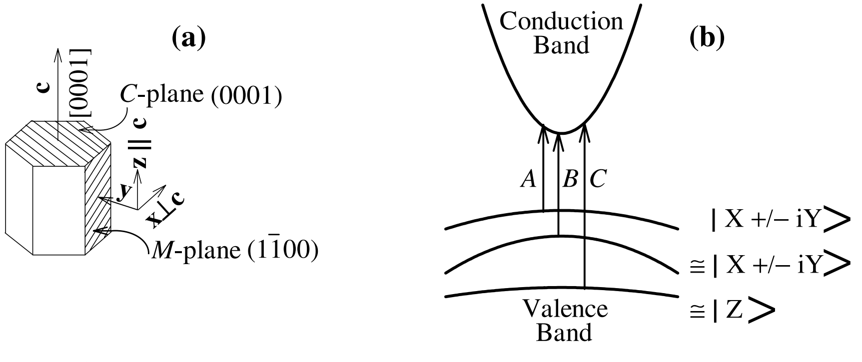

Fig. 1. a) Wurtzite GaN unit cell showing the choice of coordinates. b) Schematic diagram of the EBS of unstrained GaN (estimated exciton transition energies are $E_A = 3.410$, $E_B = 3.417$, and $E_C = 3.44$ eV at 295 K) with the corresponding valence-band wavefunction symmetry

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EBS modifications due to strain in the *M* plane [2, 3] can be quite different from the well studied case of strain in the *C* plane [4, 5].

In this paper we study a *C*-plane and an *M*-plane film with polarized photoreflectance (PR) spectroscopy, wherein we rotate the electric-field vector **E** of the linearly polarized probe beam in the film plane. We also determine theoretically the influence of an arbitrary strain in the *M* plane and the *C* plane on the EBS of GaN and its consequences for the oscillator strengths of optical transitions at the fundamental band gap. We show that the energies and polarization properties of the observed spectral features can be explained by taking the in-plane strain into account. We discuss the implications of our results for *M*-plane-based optoelectronic devices.

Experimental Details The *M*-plane GaN film was grown by rf plasma-assisted molecular-beam epitaxy (MBE) on a γ-LiAlO$_2$(100) substrate [6]. High resolution triple-axis X-ray diffraction (XRD) and Raman spectroscopy were used to verify the *M*-plane orientation of the film and its single phase nature. XRD measurements reveal biaxial compressive strain (with strain components ϵ_{xx} and $\epsilon_{zz} < 0$) in the film with an out-of-plane dilatation $\epsilon_{yy} = 0.29\%$. The *C*-plane GaN film was grown by reactive MBE on a 6H-SiC (0001) substrate [7] and is under biaxial tensile strain (ϵ_{xx} and $\epsilon_{yy} > 0$) with an out-of-plane contraction $\epsilon_{zz} = -0.08\%$. The n-type doping level in both films was about 5×10^{17} cm^{-3}.

In the PR measurements, a He–Cd laser (3.815 eV) was used as the pump beam. The probe beam with an angle of incidence of about 10° was obtained by dispersing the output of a 75 W Xe lamp using a 0.64 m monochromator (energy band pass about 4 meV) and was linearly polarized with a Glan-Taylor prism. The detector was a UV-enhanced silicon photodiode. In the measurements, the orientation of **E** (i.e., the polarization angle ϕ) was changed in the plane of the film. In the *C*-plane film, we always have $\mathbf{E} \perp \mathbf{c}$, while in the *M*-plane film we can achieve both $\mathbf{E} \perp \mathbf{c}$ and **E** (parallel $\|$) to **c**.

Polarized Photoreflectance Spectra Figure 2a shows the PR spectrum of the *C*-plane film recorded at 295 K for three values of ϕ, where $\phi = 0°$ refers to an arbitrarily chosen direction in the *C* plane. The observed resonant feature in each spectrum can be resolved into two distinct ones at 5 K (not shown here). Therefore, even at 295 K a two oscillator model was applied to fit the spectrum using Aspnes's lineshape function with modified fitting parameters [8]. The exponent *m* in the lineshape function was taken to be 3 to represent a Gaussian-broadened excitonic transition. The two transition energies obtained from fitting, labeled $E^{\mathrm{av}}_{\mathrm{T}_1,\mathrm{T}_2}$ and E_{T_3}, are independent of ϕ with values 3.402 and 3.42 eV, respectively. These spectra together with the polar plot in Fig. 2b, which shows the variation of the PR signal with ϕ at 3.4 eV, demonstrate the absence of any significant in-plane polarization anisotropy in this film.

Figure 3a shows the PR spectrum of the *M*-plane film for $\phi = 90°$ ($\mathbf{E} \perp \mathbf{c}$), $\phi = 45°$, and $\phi = 0°$ ($\mathbf{E} \parallel \mathbf{c}$) at 295 K. For each polarization, the spectrum consists of a single feature (even at 5 K) and therefore a single-oscillator lineshape function was used to fit each spectrum. Unlike in the case of the *C*-plane film, the spectral feature in the *M*-plane film shows a polarization-dependent energy shift. The transition energies for the three polarizations have been labelled E_{T_1}, E_{arbit}, and E_{T_2} with values 3.428, 3.483, and 3.468 eV, respectively. Thus, the PR spectrum of the *M*-plane film depends critically on ϕ. If the spectra for $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \perp \mathbf{c}$ are the only two possible independent

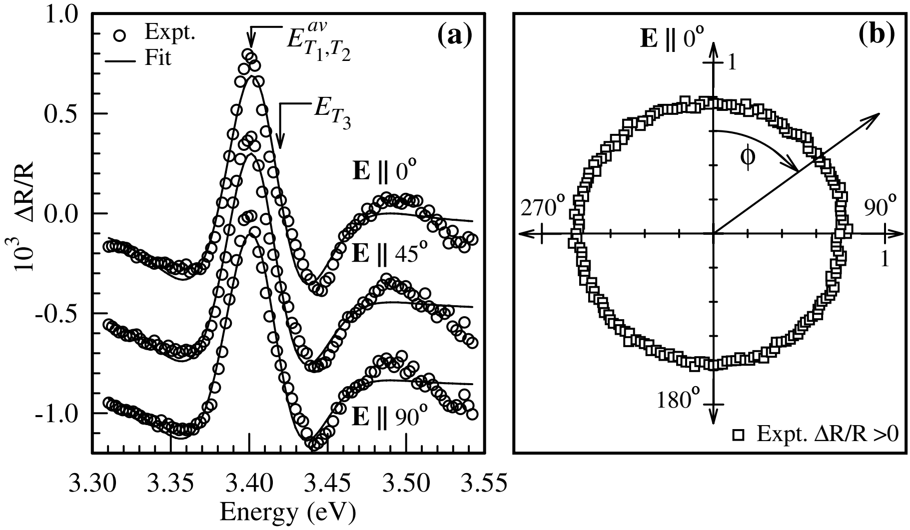

Fig. 2. a) PR spectra at 295 K of the *C*-plane GaN film on SiC for three in-plane polarizations of the probe beam. 0° refers to an arbitrarily chosen direction in the *C* plane. The plots for 45° and 90° have been shifted by -4×10^{-4} and -8×10^{-4}, respectively, for clarity. b) Polar plot showing the variation of the PR signal strength in units of 10^{-3} with the polarization angle ϕ of the probe beam at a fixed energy of 3.4 eV

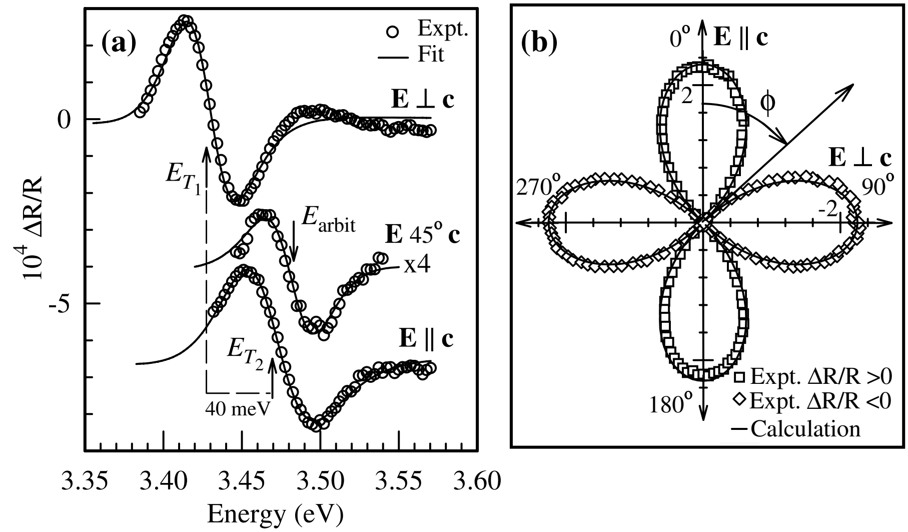

Fig. 3. a) PR spectra at 295 K of the *M*-plane GaN film on $LiAlO_2$ for three in-plane polarizations of the probe beam as indicated. The plots for **E** 45° **c** and **E** ∥ **c** have been shifted by -4×10^{-4} and -7×10^{-4}, respectively, for clarity. b) Polar plot showing the variation of the PR signal strength in units of 10^{-4} with the polarization angle ϕ of the probe beam at a fixed energy of 3.45 eV

lineshapes, the lineshape for any other ϕ can be approximated by

$$\frac{\Delta R}{R}(E,\phi) = \frac{\Delta R_{\parallel}}{R_{\parallel}}(E)\cos^2(\phi) + \frac{\Delta R_{\perp}}{R_{\perp}}(E)\sin^2(\phi)\,, \tag{1}$$

where $\Delta R_{\parallel}/R_{\parallel}$ ($\Delta R_{\perp}/R_{\perp}$) represents the lineshape measured for $\phi = 0^\circ$ ($\phi = 90^\circ$) with the assumption $R_{\parallel} \approx R_{\perp}$. This is verified in Fig. 3b, which shows that the variation of the PR signal with ϕ (at 3.45 eV) matches quite well the calculated variation based on Eq. (1) using $\Delta R_{\parallel}/R_{\parallel} = 2.3 \times 10^{-4}$ and $\Delta R_{\perp}/R_{\perp} = -2.25 \times 10^{-4}$.

Calculated Strain Dependence of the Electronic Band Structure The influence of strain on the conduction-band (CB) and the valence-band (VB) states at the Brillouin-zone centre (BZC) was calculated using the $\mathbf{k}\cdot\mathbf{p}$ method. For the three VB states closely spaced in energy, the Bir–Pikus Hamiltonian (6×6 matrix) was diagonalized [9, 10]. We used the following deformation potentials under a quasi-cubic approximation: $D^{\mathrm{CB}} = -44.5$ eV, $D_1^{\mathrm{VB}} = -41.4$ eV, $D_2^{\mathrm{VB}} = -33.3$ eV, $D_3^{\mathrm{VB}} = 8.2$ eV, $D_4^{\mathrm{VB}} = -4.1$ eV, and $D_5^{\mathrm{VB}} = -4.7$ eV [5, 10]. The other EBS parameters Δ_1 and Δ_2 as well as the exciton binding energy E_{ex} were taken to be 22, 5, and 26 meV, respectively [5]. We used the elastic constants from Ref. [11]. The momentum matrix elements $\parallel$ and $\perp$ to $\mathbf{z}$ are equal according to Ref. [12].

Strain significantly modifies the symmetry of the VB states, so it is no longer possible to describe the transitions in terms of the A, B, and C exciton transitions (cf. Fig. 1b) of unstrained GaN. We adopt the nomenclature T_1, T_2, and T_3 in the order of increasing transition energy. Figures 4a and b show the calculated energies E_{T_i} ($i = 1-3$) as well as the difference $E_{T_2} - E_{T_1}$ [4 contour plots] and the oscillator strength components f_β ($\beta = x, y, z$) [9 gray scale plots], respectively, for an arbitrary C-plane strain in the

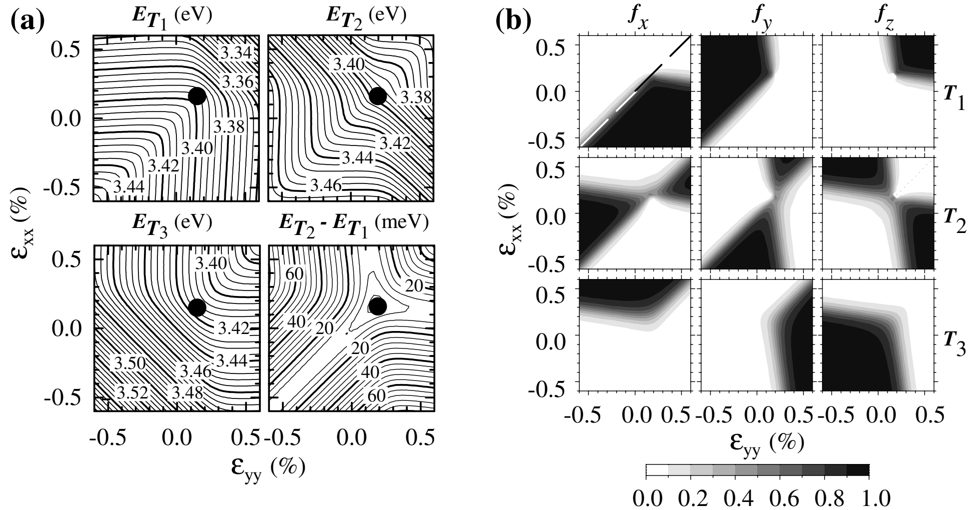

Fig. 4. a) Energies E_{T_i} of the three transitions at the fundamental band gap of GaN at 295 K as well as the difference $E_{T_2} - E_{T_1}$ and b) the oscillator strength components f_β ($\beta = x, y, z$) as a function of C-plane strain. The dashed line represents $\epsilon_{xx} = \epsilon_{yy}$. The dots in a) mark the strain in the C-plane film obtained from XRD measurements

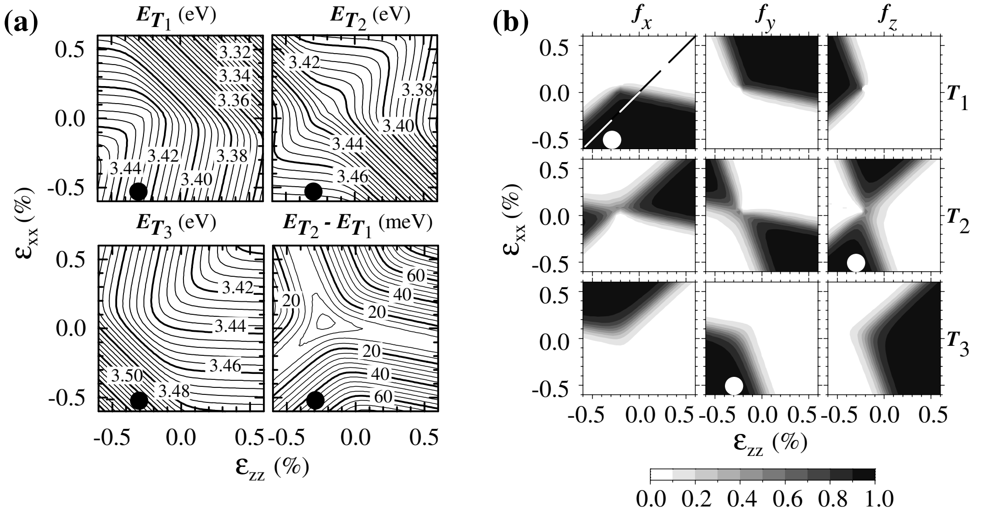

Fig. 5. a) Energies E_{T_i} of the three transitions at the fundamental band gap of GaN at 295 K as well as the difference $E_{\mathrm{T}_2} - E_{\mathrm{T}_1}$ and b) the oscillator strength components f_β ($\beta = x, y, z$) as a function of M-plane strain. The dashed line represents $\epsilon_{xx} = \epsilon_{zz}$. The dots mark the strain in the M-plane film obtained from XRD and PR measurements

range $|\epsilon_{xx}|$ and $|\epsilon_{yy}| \leq 0.6\%$. We see that f_y can be obtained by rotating f_x by 180° about the line $\epsilon_{xx} = \epsilon_{yy}$, along this line $f_y = f_x$. Thus, for isotropic C-plane strain ($\epsilon_{xx} = \epsilon_{yy}$), one does not expect any significant in-plane (x–y plane) polarization anisotropy. However, for anisotropic C-plane strain [13, 14], which breaks the crystal symmetry in the x–y plane, one can achieve a polarization anisotropy in the x–y plane, since then $f_y \neq f_x$ (cf. Fig. 4b). For a strain $\epsilon_{xx} = \epsilon_{yy} \approx 0.17\%$ the energy splitting $E_{\mathrm{T}_2} - E_{\mathrm{T}_1} \approx 0$. At this point, the in-plane strain nullifies the combined crystal-field and spin-orbit splitting of the top two VBs and thus mimics the EBS of an unstrained zincblende crystal at the BZC.

Figures 5a and b show the results of the EBS calculations for M-plane strain with $|\epsilon_{xx}|$ and $|\epsilon_{zz}| \leq 0.6\%$. In this case, f_z cannot be obtained by rotating f_x by 180° about the line $\epsilon_{xx} = \epsilon_{zz}$, along this line $f_x \neq f_z$. Therefore in unstrained or isotropically strained ($\epsilon_{xx} = \epsilon_{zz}$) M-plane films there can be a large in-plane (x–z plane) polarization anisotropy. Note that for M-plane biaxial compressive strain with ϵ_{xx} and $\epsilon_{zz} - 0.2\%$, the transitions T_1, T_2 and T_3 are predominantly x, z, and y polarized, respectively. For M-plane strain, $E_{\mathrm{T}_2} - E_{\mathrm{T}_1} \approx 0$, when $\epsilon_{xx} \approx 0.04\%$ and $\epsilon_{zz} \approx -0.24\%$.

Discussion of Results The C-plane GaN film is grown on a 6H-SiC(0001) substrate, which has hexagonal symmetry as the film. Therefore, the film experiences isotropic in-plane strain. This, according to the above theoretical results, explains the absence of any significant in-plane polarization anisotropy in the PR spectrum of this film. We identify the measured energy $E^{\mathrm{av}}_{\mathrm{T}_1,\mathrm{T}_2}$ as the average energy of the T_1 and T_2 transitions. The measured energy E_{T_3} is associated with the T_3 transition. For isotropic C-plane strain with $\epsilon_{zz} = -0.08\%$ (XRD measurement), the value of $\epsilon_{xx} = \epsilon_{yy} = 0.15\%$ is uniquely determined [5]. For such strain, the calculated energy separation $E_{\mathrm{T}_2} - E_{\mathrm{T}_1} < 2$ meV. Therefore, even at low temperatures, T_1 and T_2 cannot be resolved, and we see only two features in the 5 K spectrum, one due to the unresolved T_1 and T_2 and the other one due to the T_3 transition. When compared with the symmetry properties of the A, B, and C exciton transitions, T_1 corresponds to the A exciton, T_2 has 58% (42%) C-(B-) like character and T_3 has 58% (42%) B-(C-) like character.

In the *M*-plane film, noting the calculated polarization characteristics of T_1 and T_2 for overall compressive strain (cf. Fig. 5b), we identify the feature seen for $\mathbf{E} \perp \mathbf{c}$ with T_1 and that for $\mathbf{E} \parallel \mathbf{c}$ with T_2. The transition T_3 cannot be observed under normal incidence, since it is predominantly *y* polarized. Thus, the effective optical band gap of the *M*-plane film increases by 40 meV at 295 K, when **E** is rotated by 90° from $\mathbf{E} \perp \mathbf{c}$ to $\mathbf{E} \parallel \mathbf{c}$. The lattice constants and thermal expansion coefficients of GaN along **x** and **y** are different relative to the ones of $LiAlO_2$, therefore one expects an anisotropic in-plane strain in the *M*-plane film. Hence, it is not possible to determine ϵ_{xx} and ϵ_{zz} from the dilatation ϵ_{yy} alone, which is obtained through XRD measurements. We determined ϵ_{xx} and ϵ_{zz} as follows. Comparing the experimental E_{T_1} and E_{T_2} values with the calculations, we can estimate the in-plane strain and therefore ϵ_{yy}. Varying only the deformation potential D_5, we are able to obtain the experimental value of $\epsilon_{yy} = 0.29\%$ from $\epsilon_{xx} = -0.56\%$ and $\epsilon_{zz} = -0.31\%$ [10].

Normally, when PR measurements are used for the characterization of III–V semiconductors and alloys with (001) zincblende or *C*-plane wurtzite structures, the polarization of the probe beam is not an issue. However, with *M*-plane wurtzite nitrides, using an unpolarized probe beam or one that is polarized at an angle different from $\phi = 0^\circ$ or 90°, the resulting spectrum would be a weighted sum [according to Eq. (1)] of the two spectra for $\phi = 0^\circ$ and 90°, for example the spectrum for $\phi = 45^\circ$ in Fig. 3a. From such a spectrum no meaningful EBS parameter can be extracted so that the measured transition energy $E_{\text{arbit}} = 3.483$ eV is of no significance.

The most significant property of the strained *M*-plane film is the large, polarization-dependent change of its effective optical band gap [15]. The wavelength range, over which the resultant in-plane polarization anisotropy in the absorption coefficient exists, can be further enhanced by an appropriate choice of the in-plane strain. This has implications for lasers, if *M*-plane GaN active layers are used with in-plane compressive strain ϵ_{xx} and $\epsilon_{zz} \lesssim -0.2\%$. The larger separation (compared to *C*-plane strain) between the top two VB states lowers the density of states at the VB edge and will lead to a lower transparent carrier density. With T_1 being completely *x* polarized, an edge-emitting laser will work efficiently in transverse electric mode, when the cavity is oriented $\parallel$ to **c**. The problem of fixing the output polarization direction of small area vertical-cavity surface-emitting lasers will also no longer exist, since for GaN-active layers under such compressive *M*-plane strain the output polarization will always be $\perp$ to **c**.

Summary We have experimentally shown that, in contrast to *C*-plane GaN films, the PR spectrum of *M*-plane GaN films has a large in-plane polarization anisotropy. These results are explained by comparison with theoretical EBS calculations, which reveal the dominant role of in-plane strain, especially anisotropic in-plane strain, in determining the polarization characteristics of the interband transitions. Our work suggests that it is crucial to take into account the in-plane strain-induced EBS changes in the design of optoelectronic devices based on *M*-plane GaN films.

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