Polarized photoreflectance spectroscopy of strained *A*-plane GaN films on *R*-plane sapphire

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We have investigated a $[11\bar{2}0]$-oriented *A*-plane GaN film on *R*-plane sapphire, where the *c* axis of GaN lies in the film plane, by polarized photoreflectance (PR) spectroscopy. Near the fundamental energy gap of GaN, the PR spectrum with the probe light polarized perpendicular to the *c* axis exhibits one feature corresponding to a single transition labeled T_1. For a polarization parallel to *c* axis, two different features labeled T_2 and T_3 are observed at higher energies than the transition T_1. In order to explain the origin of these three features, we compare the measured energies with calculations of the transition energies and oscillator strengths of the three band-to-band transitions of GaN near its fundamental gap for an anisotropic in-plane strain in the *A* plane. The analysis shows that the observed transition energies and polarization properties of the three transitions can be explained by the presence of an overall compressive, anisotropic in-plane strain in the film. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1968424]

Light-emitting diodes and lasers based on group-III nitrides are usually realized with *C*-plane-oriented multiple quantum wells (MQWs) exhibiting the wurtzite crystal structure, the unit cell of which is shown in Fig. 1(a). The *c* axis for *C*-plane films and MQW structures is parallel to the growth direction. Due to the presence of large pyroelectric and piezoelectric effects, an electric fields on the order of 1 MV/cm can exist in *C*-plane MQWs along the *c* axis, which in this case is the direction of carrier confinement.[1] This electric field spatially separates the electrons and holes in the light-emitting QWs, thereby reducing the radiative efficiency and blueshifting the emission wavelength with increasing driving current. A solution to this problem is the realization of devices with MQW structures, where the *c* axis lies in the growth plane, i.e., using nonpolar orientations. The absence of internal electric fields in such structures was first demonstrated using *M*-plane MQWs on LiAlO$_2$ (Ref. 2) and more recently with *A*-plane MQWs on *R*-plane sapphire.[3,4]

The electronic band structure (EBS) and therefore the device performance of group-III-nitride MQWs crucially depend on the strain state of the structure. This aspect has been extensively studied for *C*-plane GaN films and GaN/(Al,Ga)N MQWs under isotropic strain.[5–9] For films and MQWs with a nonpolar orientation, where the *c* axis lies in the growth plane, the in-plane strain becomes anisotropic, i.e., the strain is different for the directions parallel (‖) and perpendicular (⊥) to the *c* axis. This is due to the anisotropy in the lattice mismatch and difference in the thermal-expansion coefficients of the materials for the film or MQW and the substrate. This has been demonstrated theoretically and experimentally for *M*-plane films.[10,11] The calculations

show that the most prominent effect of the anisotropic strain is a change in the optical polarization selection rules, which critically depend on the actual values of the in-plane strain.

We have investigated the influence of anisotropic strain in the *A* plane on the EBS of GaN. Using photoreflectance (PR) spectroscopy with a polarized probe beam, we have determined the energies and polarization properties of the three transitions in the vicinity of the energy gap of a strained *A*-plane GaN film. By comparing the measured transition energies and polarization directions with the respective results of band-structure calculations, we can identify the three observed transitions and explain their polarization properties. Finally, we discuss the implications of these polarization properties for device applications.

The *A*-plane GaN film was grown by metal-organic vapor-phase epitaxy on *R*-plane sapphire. The sample consists of a 0.2-μm AlN nucleation layer followed by a 1-μm-thick, undoped GaN film.[12] The *A*-plane description of the growth conditions and the postgrowth characterization of such films can be found in Ref. 12. The *A*-plane orientation of the GaN film was verified using high-resolution triple-axis
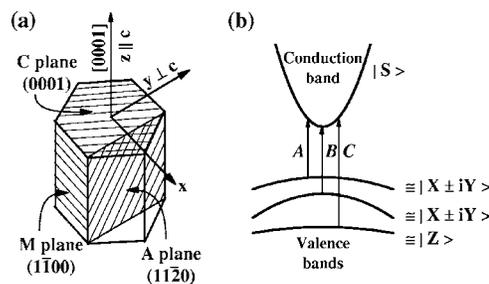

FIG. 1. (a) The wurtzite unit cell of GaN showing the *A*, *M*, and *C* planes and the choice of coordinates. (b) Schematic EBS of unstrained GaN, showing the CB and VB wave-function symmetries at the center of the Brillouin zone. For zero strain, the transitions T_1, T_2, and T_3 correspond to the *A*, *B*, and *C* excitonic transitions, respectively.

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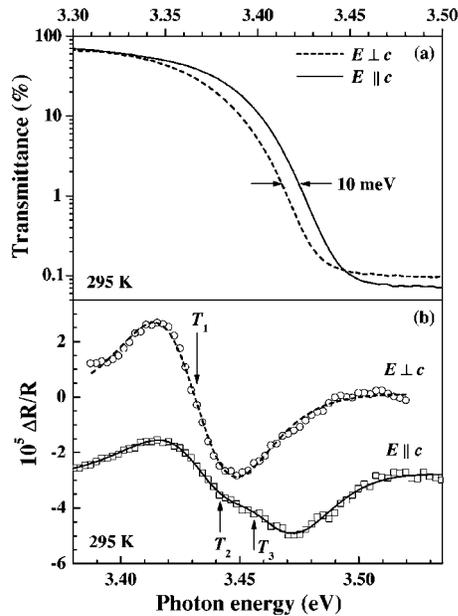[b)]Electronic mail: htgrahn@pdi-berlin.de

FIG. 2. Measured (a) transmittance and (b) PR spectra (circles and squares) of the A-plane GaN film for $E \perp c$ and $E \| c$ measured at 295 K. In (b), the curve for $E \| c$ is shifted by -3×10^{-5} for clarity. The dashed line for $E \perp c$ in (b) represents a fit to a single transition with Aspnes' line-shape function at 3.432 eV, which corresponds to T_1. The solid line for $E \| c$ is obtained by fitting two transitions with Aspnes' line-shape functions at energies 3.441 and 3.457 eV, which correspond to transitions T_2 and T_3.

x-ray diffraction (XRD) and Raman spectroscopy. The room-temperature XRD measurements show that the GaN film has an overall in-plane biaxial compressive strain with an out-of-plane dilatation $\epsilon_{xx} = 0.09\%$, where x, y, and z are parallel to the $[11\bar{2}0]$, $[\bar{1}100]$, and $[0001]$ directions, respectively. A compressive in-plane strain is expected in the GaN film due to the lattice mismatch in the y and z directions of -2.4% and -3.9%, respectively, relative to AlN and -16% and -1%,[13] respectively, relative to the sapphire substrate. However, due to the large lattice mismatch, the AlN and GaN films are probably relaxed at the growth temperature so that the strain is most likely due to the mismatch in thermal-expansion coefficients. For the PR measurements, the pump beam was obtained from a He–Cd laser at 325 nm (3.812 eV), while the probe beam was derived by dispersing the light from a 75-W Xe lamp with a 0.64-m monochromator and by polarizing it with a Glan-Taylor prism.

The polarized transmittance and PR spectra of the A-plane GaN film recorded at 295 K are shown in Figs. 2(a) and 2(b), respectively, for the electric vector E oriented $\|$ and $\perp$ to c. In Fig. 2(a), the transmittance spectrum for $E \| c$ is clearly shifted to higher energies by about 10 meV in comparison with the spectrum for $E \perp c$. The PR spectra in Fig. 2(b) have been fitted using Aspnes' line-shape function[14] with Gaussian-broadened excitonic transitions, which are expected at higher temperatures. The PR spectrum for $E \perp c$ in Fig. 2(b) can be fitted with a single feature. If we fit the PR spectrum for $E \| c$ to a single transition, the linewidth is about 60% larger than the one for the transition observed for $E \perp c$. At the same time, in order to obtain a good fit to the observed PR spectrum for $E \| c$, we have to use two transitions with energies higher than the one for $E \perp c$. We therefore

conclude that the PR spectrum for $E \perp c$ can be described by a single transition, while for $E \| c$ two transitions are necessary. The obtained transition energies are 3.432 eV for $E \perp c$ and 3.441 as well as 3.457 eV for $E \| c$ with an error of ± 3 meV. Thus, the effective energy gap of the A-plane GaN film increases by about 9 meV, when the polarization of the light is rotated by 90° from $E \perp c$ to $E \| c$. This shift agrees with the one observed in the transmittance spectra.

Unstrained wurtzite GaN has three closely spaced valence bands (VBs) [cf. Fig. 1(b)]. Excitons involving electrons in the conduction band (CB) and holes in these VBs are referred to as A, B, and C excitons with transition energies of 3.410, 3.418, and 3.440 eV, respectively, at 295 K.[7] The VBs are made of p orbitals with wave functions mainly of symmetry-type $|X \pm iY\rangle$ for A and B, while they are of $|Z\rangle$-type for C [cf. Fig. 1(b)]. In order to calculate the transition energies of the anisotropically strained A-plane GaN film, we use the $\mathbf{k} \cdot \mathbf{p}$ approximation to determine the EBS. Since the Bir-Pikus Hamiltonian[15] for wurtzite GaN under biaxial strain is symmetric with respect to the interchange of x and y, we should be able to switch from M-plane to A-plane GaN by exchanging x with y everywhere and vice versa. We have checked this symmetry by extending the calculations of the transition energies for M-plane GaN presented in Ref. 11 to anisotropically strained A-plane GaN films.[16] Since the previous calculations were performed to determine the transition energies at low temperatures, we have subtracted 70 meV, which was determined by averaging the Varshni parameters of several references in Ref. 9, from the low-temperature energies to obtain the corresponding room-temperature energies. Except for the energy shift, the strain dependence of the transition energies for A-plane GaN films at 295 K looks therefore basically the same as for M-plane GaN films at 5 K, as shown in Ref. 11. The anisotropic strain mixes the VB states, so that it is no longer possible to describe the three transitions in terms of the A, B, and C excitons. We therefore adopt the nomenclature described in Ref. 11, where the transitions are labeled T_1, T_2, and T_3 in order of increasing energies E_1, E_2, and E_3, respectively.

Figure 3(a) shows the variation of the out-of-plane strain ϵ_{xx} as a function of the in-plane strain components ϵ_{yy} and ϵ_{zz}, while Figs. 3(b)–3(d) present the calculated energies vs ϵ_{yy} and ϵ_{zz} for the three transitions T_1, T_2, and T_3, respectively. In Figs. 3(b)–3(d), the contours marked by the dotted lines correspond to the measured PR transition energies. Using Fig. 3, we interpret the PR results in the following way. If we identify the PR feature lowest in energy at 3.432 eV with the T_1 transition, we derive from Figs. 3(a) and 3(b) that the value of our measured out-of-plane strain component $\epsilon_{xx} = 0.09\%$ intersects the energy $E_1 = 3.432$ eV for the two in-plane strain coordinates $\epsilon_{yy} = 0.05\%$ and $\epsilon_{zz} = -0.39\%$ as well as $\epsilon_{yy} = -0.05\%$ and $\epsilon_{zz} = -0.26\%$. For the first set of strain coordinates, the calculated energies for the transitions T_2 and T_3 are $E_2 = 3.444$ eV and $E_3 = 3.457$ eV, while for the second set they are $E_2 = 3.438$ eV and $E_3 = 3.454$ eV. From these energy values alone, we cannot distinguish between the two sets of in-plane strain components.
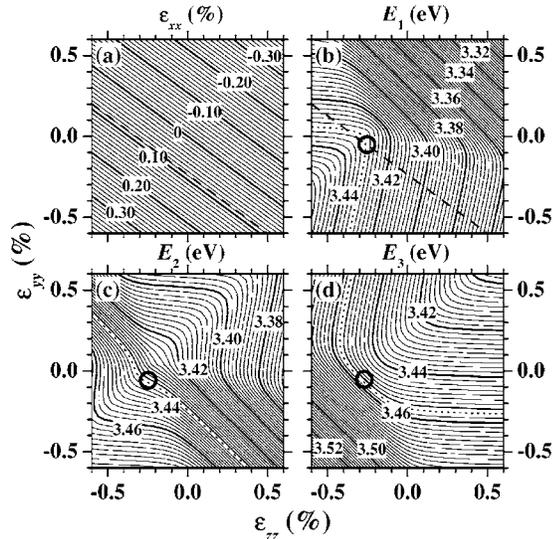
FIG. 3. Contour plots showing the variation (a) in the out-of-plane strain ϵ_{xx}, (b) in the energy E_1 of the transition T_1, (c) in the energy E_2 of the transition T_2, and (d) in the energy E_3 of the transition T_3 as a function of in-plane strain ϵ_{yy} and ϵ_{zz} in the A plane of GaN. The dashed lines in (a) and (b) trace the contour of $\epsilon_{xx}=0.09\%$. The dotted lines in (b), (c), and (d) indicate the measured transition energies. The circles in (b), (c), and (d) mark the in-plane strain coordinates $\epsilon_{yy}=-0.05\%$ and $\epsilon_{zz}=-0.26\%$, which are identified as being present in the investigated film.

FIG. 4. Relative magnitudes $f_{i\beta}$ ($i=1,2,3$ and $\beta=x,y,z$) representing the oscillator strengths for the three transitions T_1, T_2, and T_3 as a function of in-plane strain ϵ_{yy} and ϵ_{zz} for A-plane GaN. T_1 (T_3) is defined as the transition with the lowest (highest) transition energy for a given in-plane strain value. The circles mark the in-plane strain coordinates $\epsilon_{yy}=-0.05\%$ and $\epsilon_{zz}=-0.26\%$, which are identified as being present in the investigated film.

In order to unambiguously identify the strain values, we have to determine the oscillator strength of the three transitions T_1, T_2, and T_3. The relative oscillator strength components $f_{i\beta}$, where $i=1,2,3$ correspond to the three transitions T_1, T_2, and T_3, while $\beta=x,y,z$ are the three components along x, y, and z directions that determine the optical polarization selection rules, are plotted in Fig. 4 as a function of A-plane strain. The energy and polarization properties of T_1, T_2, and T_3 at zero strain are identical to the ones of the A, B, and C excitons. Note that the x polarization is not accessible with a normally incident beam on an A-plane film. These plots clearly demonstrate that only for the in-plane strain values $\epsilon_{yy}=-0.05\%$ and $\epsilon_{zz}=-0.26\%$ the oscillator strength of the T_1 transition is significant for y polarization ($E\perp c$) and negligible for z polarization ($E\|c$). In addition, the T_2 and T_3 transitions exhibit appreciable oscillator strengths only for the z polarization and not for the y polarization, in agreement with the measured polarization dependence of the PR spectra. Therefore, only the calculated transition energies together with the calculated oscillator strengths allow for an unambiguous identification of the in-plane strain components for the A-plane GaN film. Note that the exact values of the deformation potentials are still under investigation. Hence, the values of the calculated transition energies and oscillator strengths may change somewhat, but the overall strain dependence and the resulting agreement with our experimental data will not be significantly affected.

The calculated oscillator strengths shown in Fig. 4 demonstrate that for large in-plane compressive strain the T_1 transition, which is the lowest in energy, is predominantly y polarized, while for tensile strain it becomes x polarized. This property suggests that, if edge-emitting lasers are realized with A-plane GaN films in the active region, efficient la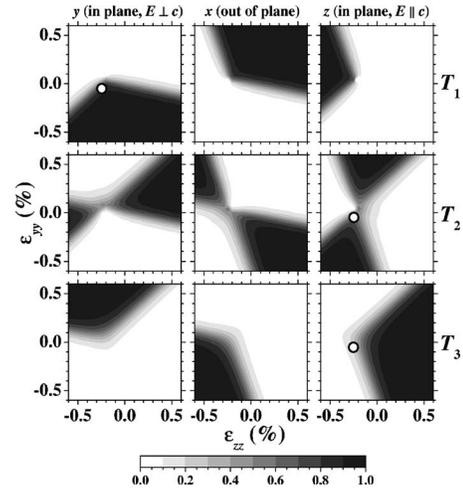sing in the transverse electric mode can occur for compressive strain, while lasing in the transverse magnetic mode would be favored for tensile strain. In the former case, it would be necessary to orient the laser cavity along the c axis, since light with $E\|y$ will not travel along the y direction. Thus, the presence of anisotropic strain in group-III-nitride devices based on A-plane films will have significant consequences for the design of optoelectronic devices.

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