

Optically detected magnetic resonance of (effective-mass) shallow acceptors in Si-doped GaN homoepitaxial layers

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Optically detected magnetic resonance (ODMR) has been performed on Si-doped GaN homoepitaxial layers grown by organometallic chemical vapor deposition on free-standing GaN templates. In addition to intense excitonic bandedge emission with narrow linewidths (<0.4 meV), these films exhibit strong shallow donor–shallow acceptor recombination at 3.27 eV. Most notably, ODMR on this photoluminescence band reveals a highly anisotropic resonance with $g_{\parallel} = 2.193 \pm 0.001$ and $g_{\perp} \sim 0$ as expected for effective-mass shallow acceptors in wurtzitic GaN from $\mathbf{k} \cdot \mathbf{p}$ theory. This previously elusive result is attributed to the much reduced dislocation density and impurity levels compared to those typically found in the widely investigated Mg-doped GaN heteroepitaxial layers. The possible chemical origin of the shallow acceptors in these homoepitaxial films will be discussed.

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I. INTRODUCTION

The nature and properties of prospective p -type group-II dopants such as Mg, Zn, and Be and group-IV elements such as C in GaN and related alloys have been topics of much interest during the last several years. The most viable impurity to emerge from this list of candidates for achieving high hole densities is Mg. However, one of the puzzles revealed from a variety of magnetic resonance studies,^{1–5} including electron paramagnetic resonance (EPR) and optically detected magnetic resonance (ODMR), of Mg-doped GaN heteroepitaxial layers grown by metal organic chemical vapor deposition or molecular beam epitaxy, is the nearly isotropic Landé g -tensor (i.e., $g_{\parallel}, g_{\perp} \sim 2$, where $\parallel$ refers to the c axis) observed for the shallow Mg acceptors. This result contrasts with the highly anisotropic g tensor (i.e., $g_{\parallel} \sim 2-4$, $g_{\perp} \sim 0$) that is expected based on symmetry arguments and the ordering of the valence band states in wurtzitic (wz) GaN from which the shallow (bound) acceptor states are derived. One model⁶ proposed to account for this anomalous behavior is the presence of random strain fields in these structures. Such strain fields may arise, for example, from the high density of dislocations ($\sim 7 \times 10^8 - 10^{10} \text{ cm}^{-2}$) typically found in GaN heteroepitaxial films grown on Al_2O_3 and SiC substrates and those generated from the large concentration of Mg impurities ($\geq 3 \times 10^{18} \text{ cm}^{-3}$) introduced to achieve high hole densities.

In this work we report on the first direct observation of the highly anisotropic g tensor that is predicted for shallow acceptors in wz-GaN. This result was obtained by ODMR on a strong shallow donor–shallow acceptor photoluminescence (PL) band observed from a high-quality Si-doped GaN homoepitaxial layer grown by metal-organic chemical vapor deposition (MOCVD) on thick, free-standing hydride vapor phase epitaxy (HVPE) GaN templates. The main characteristics of this resonance, including its g tensor ($g_{\parallel} = 2.193$,

$g_{\perp} \sim 0$) and weak intensity for field orientations near the c axis, are similar to those found for shallow acceptors in other bulk semiconductors with similar valence band ordering and crystal structure such as CdS (Ref. 7) and 6H-SiC (Refs. 8 and 9). This new feature is tentatively assigned to Si_N (Si atoms on the N lattice sites) shallow acceptors with a binding energy (E_a) of ~ 220 meV. However, we can not rule out that all or part of this signal is associated with residual C_N shallow acceptors.

II. EXPERIMENTAL DETAILS

The PL and ODMR studies were performed on $\sim 6 \mu\text{m}$ -thick Si-doped GaN layers deposited by MOCVD on HVPE-grown free-standing GaN substrates ($\sim 5 \times 5 \text{ mm}^2$). Recent x-ray diffraction, atomic force microscopy (AFM), transmission electron microscopy (TEM), PL, and Raman scattering measurements^{10–13} all indicate the high crystalline quality of these HVPE GaN substrates. In particular, this material is characterized by dislocation densities less than 10^7 cm^{-2} as revealed by TEM (Ref. 10) and AFM (Ref. 11) and exhibits strong bandedge excitonic PL with linewidths less than 0.4 meV (Refs. 13 and 14). The Ga face of the $\sim 400\text{-}\mu\text{m}$ -thick GaN template was mechanically polished and reactive ion etched prior to the chemical vapor deposition (CVD) growth. Si doping of the CVD homoepitaxial layer was achieved with $\sim 1/3$ of the Silane precursor flow employed in previous growths to give n -type films with $\sim 3 \times 10^{17} \text{ carriers/cm}^3$. Additional details of the epigrowth are provided elsewhere.¹⁵

The PL at 1.6 K was excited by the 351-nm line of an Ar^+ laser. The emission between 1.8 and 3.3 eV was analyzed by a $\frac{1}{4}$ -m double-grating spectrometer and detected by a Si photodiode. The ODMR was performed in a 24-GHz spectrometer described elsewhere¹⁶ with the sample rotated in the (1120) plane to obtain symmetry information. The ODMR was obtained with either visible bandpass or cutoff filters placed in front of the Si photodiode to separate the individual

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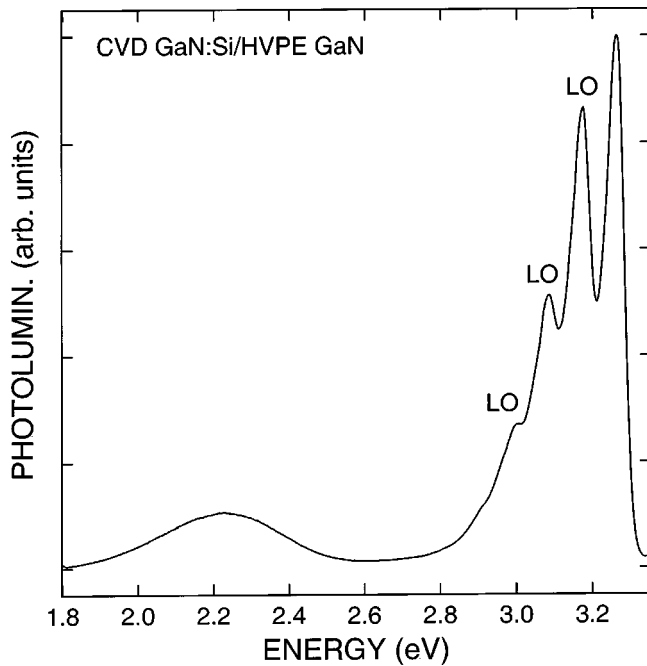


FIG. 1. PL obtained below 3.4 eV at 1.6 K from a Si-doped GaN homoepitaxial layer. The zero-phonon line (ZPL) at 3.27 eV is assigned to shallow donor–shallow acceptor recombination (LO, longitudinal optical phonon replica).

PL bands. We note that the strongest signals were found with a microwave amplitude modulation frequency of ~ 3 kHz and an excitation power density of ~ 1 W/cm².

III. RESULTS AND DISCUSSION

Several recombination bands were observed at low temperature from the Si-doped GaN homoepitaxial layer. The dominant emission at 3.472 eV found from high-resolution PL studies (reported elsewhere¹⁷) is assigned to excitons bound to shallow Si donors. The reduced linewidth of this emission (≤ 0.4 meV) compared to that typically observed¹⁷ for GaN grown on sapphire or SiC with similar Si doping levels indicates the high-crystalline quality of these homoepitaxial films.¹⁸ The PL spectrum below 3.4 eV from this layer is shown in Fig. 1. Very strong recombination at ~ 3.27 eV and a series of longitudinal optical phonon replicas are observed. In addition, this PL shifts monotonically to lower energy with decreasing excitation power density. Similar emission of varying strength has been found by several groups for nominally undoped, Si-doped, and Mg-doped GaN heteroepitaxial layers.^{15,16,19–21} Based on these reports, the 3.27-eV PL is assigned to recombination between shallow Si donors ($E_d \sim 30$ meV) and shallow acceptors ($E_a \sim 220$ meV) whose possible origin will be discussed later. The 3.27-eV shallow donor–shallow acceptor (SD–SA) PL was only *weakly* observed from a nominally undoped GaN homoepitaxial reference layer grown for this study on another piece of the free-standing HVPE substrate.

In addition to the SD–SA PL band, a broad “yellow” emission band at ~ 2.2 eV is also found as often observed to some degree of strength from highly resistive and *n*-type

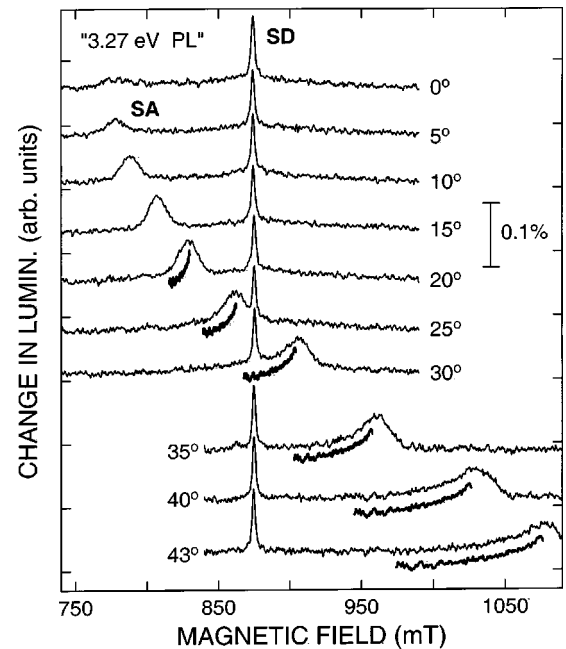


FIG. 2. ODMR spectra at 24 GHz detected on the 3.27-eV SD–SA PL for several orientations of the applied magnetic field ($\mathbf{B}$) in the (1120) plane (0° refers to the *c* axis). The thick black curves (displaced vertically for clarity) are simulations of the low-field lineshapes for resonance SA as described in the text (SD, shallow donor; SA, shallow acceptor).

(either as-grown or Si-doped) CVD heteroepitaxial layers.¹⁶ The microscopic origin of the deep center involved in this emission has been a subject of high interest during the last several years. Possible models include its association with residual C impurities, lattice defects such as Ga vacancies, and extended defects such as threading dislocations. Though not the focus in this paper, we note that ODMR was performed on the 2.2-eV band from the Si-doped GaN homoepitaxial layer. Unfortunately, as in the case for the heteroepitaxial films characterized by much higher dislocation densities,¹⁶ resolved electron–nuclear hyperfine structure that could shed light on its identity was not observed for the deep center with $g_{\parallel} = 1.989$ and $g_{\perp} = 1.992$ revealed from ODMR on this “yellow” emission.

The ODMR obtained on the 3.27-eV SD–SA recombination from the Si-doped GaN homoepitaxial layer for several orientations of the applied magnetic field ($\mathbf{B}$) is shown in Fig. 2. Two luminescence-increasing signals are observed. The first (labeled SD) is sharp (full width at half-maximum of ~ 3 mT) with $g_{\parallel} = 1.952$ and $g_{\perp} = 1.949$. This g tensor has been established as a “fingerprint” for (effective-mass) shallow donors in GaN from previous magnetic resonance work^{16,22} on *n*-type films (as grown and Si doped). Thus, the feature is assigned to Si donors on the Ga lattice sites although residual O shallow donors on the N sites may also contribute. In particular, it is not expected that a small difference in g values can be easily resolved for O and Si shallow donors in GaN based on the similar binding energies²³ and the broad magnetic resonance linewidths (due to the high abundance of nearby host lattice nuclei with finite spin) typically observed for point defects in III–V semiconductors.

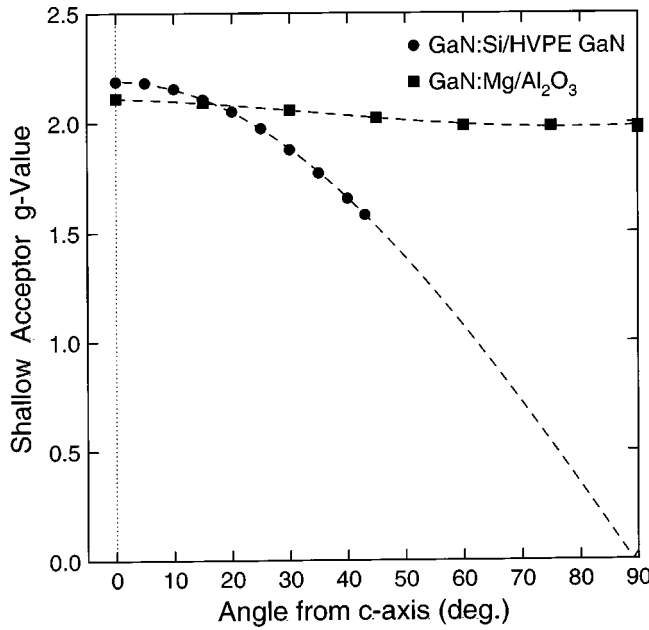


FIG. 3. g values of ODMR feature SA (circles) and those found previously for Mg shallow acceptors in Mg-doped GaN heteroepitaxial layers (squares) as a function of the angle (θ) between $\mathbf{B}$ and the c axis. The dashed curves are fits to the data [see Eq. (1)].

The second resonance (labeled SA) exhibits several distinct characteristics. First, the intensity of the line is very weak for $\mathbf{B}$ within 5° of the c axis and increases monotonically with increasing θ up to approximately 30° , where θ refers to the angle between the applied magnetic field ($\mathbf{B}$) and the c axis. Second, most notably, the peak shifts rapidly to higher field as $\mathbf{B}$ is rotated away from the c axis; especially for $\theta > 20^\circ$. We note that the resonance could not be tracked for $\theta > 43^\circ$ because B_{max} was 1100 mT for this study. Third, a symmetric Gaussian line shape with a full width at half maximum (FWHM) of ~ 15 mT is observed for the line with $\theta < 25^\circ$. However, a pronounced asymmetric broadening to the low-field side of the peaks is found for $\theta > 25^\circ$.

A plot of the g values (filled circles) as a function of θ

determined from the peaks of resonance SA is shown in Fig. 3. The g values (filled squares) reported previously¹ for Mg shallow acceptors by ODMR on the 3.27-eV SD-SA PL from Mg-doped GaN heteroepitaxial layers are also shown for comparison. Fits to these data (dashed curves) were made with the usual expression for g tensors in the case of axial symmetry:

$$g(\theta) = (g_{\parallel}^2 \cos^2 \theta + g_{\perp}^2 \sin^2 \theta)^{1/2}, \quad (1)$$

where $g_{\parallel}$ and $g_{\perp}$ are the g values with $\mathbf{B}$ aligned parallel and perpendicular to the c axis, respectively. Good fits are obtained with $g_{\parallel} = 2.193 \pm 0.001$ and $g_{\perp} \sim 0$ for resonance SA in contrast with the nearly isotropic g values ($g_{\parallel} = 2.105 \pm 0.004$ and $g_{\perp} \sim 1.970 \pm 0.005$) for Mg shallow acceptors in the GaN:Mg/Al₂O₃ films. Most notably, the highly anisotropic g tensor observed for feature SA, described by $g(\theta) = g_{\parallel}(\cos \theta)$, is predicted from effective-mass theory^{6,24} for shallow acceptors in wurtzitic GaN where the ground state, from symmetry arguments, reflects the character of the [$J = 3/2$, $m_J = \pm 3/2$] (top) valence band edge. As shown in Table I, similar highly anisotropic g tensors have been observed via ODMR for shallow acceptors in bulk CdS (Ref. 7) and 6H-SiC (Refs. 8 and 9), semiconductors with similar valence band ordering and crystal structure to that of wz-GaN. Thus, based on the PL and magnetic resonance properties, resonance SA is assigned to effective-mass (EM) shallow acceptors. This result is the first direct observation of the highly anisotropic g -tensor expected for EM acceptors in wz-GaN. A second signature of such acceptors is the weak intensity of the ODMR with θ near 0° (as also observed from ODMR of shallow acceptors in CdS and 6H-SiC). The behavior follows from the angular dependence ($\propto \tan \theta$) of the microwave transition probability for this case with $g_{\perp} \sim 0$ as described in Ref. 7.

For cases where the valence band minima are degenerate or nearly degenerate as in wz-GaN where the heavy [$J = 3/2$, $m_J = \pm 3/2$] heavy and light [$J = 3/2$, $m_J = \pm 1/2$] hole bands are split by ~ 22 meV (Ref. 25), the properties of shallow acceptors are very sensitive to the presence of random strains. In particular, it was recently proposed⁶ that random

TABLE I. Summary of g tensors found from ODMR of shallow acceptors in several wide-band-gap semiconductor materials.

Host	Shallow acceptor	g values	Reference
Bulk 6H-SiC	Al	$g_{\parallel} = (2.325 - 2.412) \pm 0.002$, $g_{\perp} = 0$	8
	Ga	$g_{\parallel} = 2.21 - 2.27$, $g_{\perp} = 0.6 \pm 0.2$	9
Bulk CdS	Li	$g_{\parallel} = 2.829 \pm 0.007$, $0.1 < g_{\perp} < 0.4$	7
	Na	$g_{\parallel} = 2.733 \pm 0.007$, $0.1 < g_{\perp} < 0.4$	
CVD GaN:Mg/Al ₂ O ₃	Mg _{Ga}	$g_{\parallel} = 2.105 \pm 0.004$, $g_{\perp} = 1.970 \pm 0.005$	1
CVD GaN:Si/Bulk GaN	Si _N (?), C _N (?)	$g_{\parallel} = 2.193 \pm 0.001$, $g_{\perp} \sim 0$	This work

in-plane strains associated with symmetry-lowering local distortions could account for the nearly isotropic g tensors observed by several groups for Mg shallow acceptors in GaN heteroepitaxial layers doped with Mg between $\sim 3 \times 10^{18}$ and $1 \times 10^{20} \text{ cm}^{-3}$. For such samples, these distortions can arise from the local strain fields associated with the high density of dislocations ($\sim 7 \times 10^8 - 10^{10} \text{ cm}^{-2}$) and, also, from the random strain fields that are produced from the size difference between the high concentration of Mg atoms and the Ga atoms they replace.²⁶ Another possible source of distortions at the Mg sites are electric fields generated from the high concentrations ($> 10^{18} \text{ cm}^{-3}$) of charged donors and acceptors randomly distributed in these highly doped and compensated films.¹ Thus, we suggest two important factors led to the observation of the highly anisotropic SA g tensor in the present Si-doped GaN homoepitaxial layer: (1) the reduced density of dislocations ($< 10^7 \text{ cm}^{-2}$), and (2) the much lower impurity concentrations ($< 3 \times 10^{17} \text{ cm}^{-3}$). The reduced degree of random strain in this film is also evident from the narrow excitonic bandedge emission and the smaller FWHM ($\sim 15 \text{ mT}$) of the SA ODMR observed with $\theta \leq 25^\circ$ compared to that found ($\sim 25 \text{ mT}$) for the ODMR of Mg shallow acceptors in the highly Mg-doped GaN heteroepitaxial layers.¹

As noted earlier, a clear asymmetric broadening of the SA ODMR is observed with $\theta > 25^\circ$ (Ref. 27). This behavior can be understood from the presence of both “unperturbed” shallow acceptors with $g_\perp \sim 0$ and “perturbed” centers with a distribution of finite $g_\perp$ values due to residual random strain fields. This distribution was obtained by combining Eq. (1), the expression for the magnetic resonance condition [$h\nu = g(\theta)\mu_B B$], and a fixed $g_\parallel$ value of 2.193 to reproduce the asymmetric lineshape observed for the $\theta = 40^\circ$ spectrum. This simulation and those obtained for the other spectra using the *same* distribution of $g_\perp$ values (between 0 and 1) with no other adjustable parameters are shown for clarity by the vertically displaced bold curves in Fig. 2. Most notably, good simulations are found for the spectra with $\theta \geq 30^\circ$. However, the simulations are poor with $\theta < 30^\circ$. This is not surprising because the $g_\parallel^2 \cos^2 \theta$ term in Eq. (1) for $\theta < 30^\circ$ is dominant so any influence on the line shape due to a variation in $g_\perp$ is significantly reduced. We note that a variation in $g_\parallel$ of only $\pm 1\%$ can account for the FWHM of $\sim 15 \text{ mT}$ observed for the SA ODMR with $\theta < 30^\circ$.

A schematic illustration based on this magnetic resonance work and that reported in the past of the $g_\perp$ -value behavior and relative concentration of shallow acceptors in wz-GaN modeled as a function of the degree of perturbation is shown in Fig. 4. The $g_\perp$ values (dotted curve) rise rapidly from 0 to a limiting value of ~ 2 with increasing perturbation proposed to arise from random strain fields, electric fields, etc. Three cases are given. The first (a) is for a “perfect” crystal (i.e., very low dislocation densities and impurity levels) where all shallow acceptors are “unperturbed” and characterized by $g_\perp$ values of ~ 0 (as depicted by the delta function). The second (b) corresponds to a weakly perturbed crystal, similar to the present Si-doped GaN homoepitaxial film, comprised of many shallow acceptors with $g_\perp \sim 0$ and a distribution of

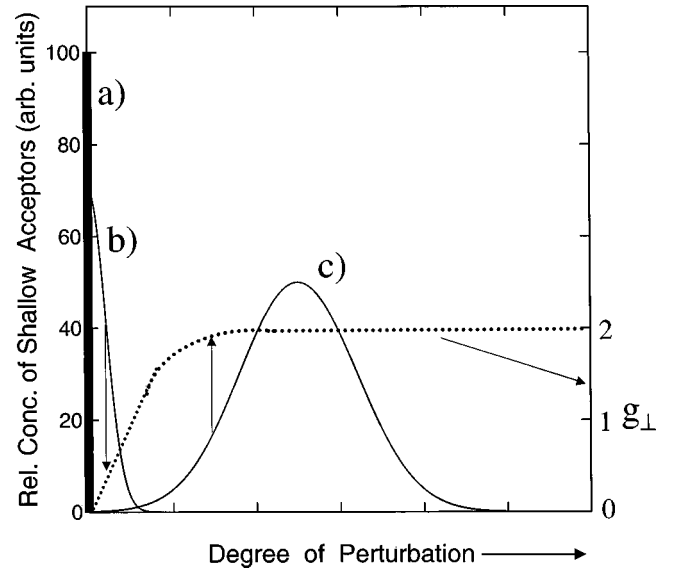


FIG. 4. A schematic representation of the $g_\perp$ value behavior (dotted curve) and relative concentration of shallow acceptors in wz-GaN modeled as a function of the degree of perturbation (e.g., in-plane random strains/electric fields, ...). Three cases are depicted: (a) a “perfect” crystal with $g_\perp \sim 0$ for all acceptors (illustrated by the delta function), (b) a weakly perturbed crystal, such as the present Si-doped GaN homoepitaxial layer, with $g_\perp \sim 0$ for many shallow acceptors and a distribution of centers with $g_\perp$ between 0 and 2, and (c) highly dislocated and heavily Mg-doped GaN heteroepitaxial layers with $g_\perp \sim 2$ for nearly all acceptors. The vertical arrows denote how the $g_\perp$ values can be read in this figure at particular points for cases (b) and (c).

“perturbed” acceptors with $g_\perp$ between 0 and 2. The final case (c) represents highly dislocated and heavily Mg-doped GaN heteroepitaxial layers where the shallow acceptors are perturbed to such degree that nearly all are characterized by $g_\perp \sim 2$.

Three possible sources are considered for the chemical identity of the shallow acceptors in this Si-doped homoepitaxial film. Mg is an obvious candidate based on its very similar binding energy ($\sim 220 \text{ meV}$) with that observed for the shallow acceptors in this sample. In addition, the $g_\parallel$ value for the EM shallow acceptors in this work is only slightly higher (~ 0.1) than that found for Mg shallow acceptors in the Mg-doped GaN heteroepitaxial layers. However, as shown in Table I, similar small differences in $g_\parallel$ values were also reported for the different EM shallow acceptor species in bulk CdS (Ref. 7) and 6H-SiC (Refs. 8 and 9). Most notably, Mg had never been introduced as a dopant source in the CVD reactor used to grow these homoepitaxial layers nor known to be a residual contaminant at the part per million levels in the source gases (TMGa, TMAI, and silane). Thus, we suggest that C_N and/or Si_N [analogous to the amphoteric nature observed for these impurities in other III-V semiconductors such as GaAs (Ref. 28)] are more likely the source of the EM acceptors. Ionization energies of $\sim 220 \text{ meV}$ have been calculated^{29,30} for both C_N and Si_N although it has also been suggested that Si_N is a deep acceptor with E_a of $\sim 1 \text{ eV}$ (Ref. 31). From size arguments³² C is more favorable than Si

to replace the N host atoms and, also, is present above the part per million level [$\sim(3-5)\times 10^{16}\text{ cm}^{-3}$] based on secondary ion mass spectroscopy (SIMS) measurements of undoped films grown in this reactor.³³ However, only a very limited number of reports³⁴ provide evidence for C as a successful *p*-type dopant in wz-GaN. Indeed, several groups suggest that C is more likely linked with deep traps such as those suggested to be involved in the ubiquitous 2.2-eV “yellow” PL band³⁵ and the current collapse phenomena often observed in CVD-grown GaN HEMTs (Ref. 33) and MESFETs (Ref. 36). Perhaps most pertinent to this work, strong SD-SA PL with zero-phonon-line (ZPL) at 3.27 eV has been reported^{15,21} for Si-doped GaN heteroepitaxial layers and suggested by one group²¹ to involve Si_N shallow acceptors from a doping study. This association is also consistent with the significant increase of the 3.27-eV SD-SA recombination observed from the present homoepitaxial GaN films after Si doping. Thus, the SA ODMR is tentatively assigned to Si_N shallow acceptors. However, it cannot be ruled out that all or part of this signal is associated with C_N shallow acceptors.

IV. SUMMARY

ODMR has been performed on strong SD-SA PL from a Si-doped GaN homoepitaxial layer grown by organometallic chemical vapor deposition on free-standing HVPE GaN tem-

plates. Most notably, a highly anisotropic resonance with $g_\parallel = 2.193 \pm 0.001$ and $g_\perp \sim 0$ is observed and assigned to effective-mass shallow acceptors from $\mathbf{k}\cdot\mathbf{p}$ theory and the ordering of the valence band states in wz-GaN. This previously elusive result via magnetic resonance techniques is attributed to the much reduced dislocation density and impurity concentrations in this homoepitaxial film compared to those in conventional Mg-doped GaN heteroepitaxial layers with doping levels greater than $3 \times 10^{18}\text{ cm}^{-3}$. The shallow acceptors are tentatively assigned to Si_N but other possible sources such as residual C impurities are not ruled out at this time. In light of these results, it would be highly interesting to perform magnetic resonance on Mg-doped GaN homoepitaxial layers as a function of doping level (e.g., $[\text{Mg}] \sim 10^{17} - 10^{20}\text{ cm}^{-3}$). In particular, such samples should allow one in a controlled way to separate the effects on the spin properties due to high dislocation densities from those due to high doping levels.

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