

Internal structure and oscillator strengths of excitons in strained α -GaN

Bernard Gil and Olivier Briot

Centre National de la Recherche Scientifique Groupe d'Etude des Semiconducteurs Université de Montpellier II, case courrier 074,
34095 Montpellier Cedex 5, France

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We calculate the excitonic exchange interaction in α -GaN, and find it to be about 2 meV. A theoretical modeling of excitons is performed, and the oscillator strengths of radiative levels are calculated as a function of strain for (0001)-grown epilayers. In particular, we find that the strength of optical transitions are extremely sensitive to the residual strain field in view of the small value of the spin-orbit interaction. Our calculation shows agreement with low-temperature reflectance investigations in GaN epilayers grown on sapphire substrates. [S0163-1829(97)10304-6]

Active research investigations are currently devoted to the group-III nitride semiconductor compounds. Modern epitaxy techniques recently allowed researchers to achieve GaN epilayers of reproducible high quality having both n and p types. Interesting devices, including light-emitting diodes, compact solid-state lasers, UV detectors and modulators as well as discrete transistors and eclectic sensors for operation in caustic conditions, now exist.¹ The race to production of such devices prevented, until recently, needed attention to the basic physical properties of the materials.

Now, for the purpose of a progression toward the design of special architectures, and a subsequent improvement of devices, we need to improve our general comprehension of the materials themselves for completion and updating of information collected some 20 years ago by the pioneers who studied GaN. In addition, other aspects have to be investigated in detail, especially those having to do with growth mechanisms in relation to the deposition process, and the recognition and treatment of topological defects. Other areas of interest include the improvement of metal-to-semiconductor contacts, and the influence of recombination mechanisms on optical properties, to cite just two examples. In this paper we develop a theoretical calculation of the excitonic oscillator strength for Γ_5 and Γ_1 excitons in wurtzitic GaN layers. The calculation, which is made at $\mathbf{k}=\mathbf{0}$, is restricted here to layers grown along the \mathbf{c} direction and includes the effect of residual biaxial stress. The electron-hole exchange interaction is estimated from a two-body calculation² which was found to account nicely for experimental results obtained on II-VI materials that are close cousins of α -GaN. We further predict that the biaxial stress effect significantly alters the oscillator strength of B and C excitons, although the effect is much less dramatic for A . Finally, a comparison is made with the experimental data, and agreement is obtained between strain-induced evolution of reflectance features and the predictions of our calculation.

Numerous papers have been written concerning excitons in wurtzite II-VI's which approximated the hexagonal symmetry as a $\langle 111 \rangle$ -distorted version of the zinc-blende crystal.³ In the current paper, the problem will be treated using more complete predictions of group theory in the context of hexagonal symmetry. The exciton Hamiltonian Ξ_{exc} is written⁴

$$\Xi_{\text{exc}} = H_{c=0} + H_{c\text{strain}} + H_{v=0} + H_{v\text{strain}} + H_{\text{exc}}, \quad (1)$$

where $H_{v=0}$ ($H_{c=0}$) are the strain-free valence (conduction) band Hamiltonians, and $H_{v\text{strain}}$ ($H_{c\text{strain}}$) account for strain-related effects on the evolution of band extrema. For the last operator on the right-hand part of Eq. (1), we write

$$H_{\text{exc}} = R^* + 1/2 \gamma \boldsymbol{\sigma}_h \cdot \boldsymbol{\sigma}_c \quad (2)$$

where R^* is the customary exciton binding energy and the last term is the crystalline exchange interaction. Operators $\boldsymbol{\sigma}_h$ and $\boldsymbol{\sigma}_c$ operate on valence-hole and conduction-electron spin functions.⁵

In the case of growth along the (0001) direction, the non-vanishing symmetrized components of the strain tensor are $e_{\Gamma_1}^{(1)} = e_{zz}$ and $e_{\Gamma_1}^{(2)} = (e_{xx} + e_{yy})$. Consequently, Eq. (1) is written as follows:

$$\begin{aligned} \Xi_{\text{exc}} = & \Delta_1 L_z^2 + \Delta_2 L_z \sigma_{vz} + \Delta_3 (L_x \sigma_{vx} + L_y \sigma_{vy}) \\ & + (a_z + b_z L_z^2) e_{\Gamma_1}^{(1)} + (a_x + b_x L_z^2) e_{\Gamma_1}^{(2)} \\ & + 1/2 \gamma \boldsymbol{\sigma}_h \cdot \boldsymbol{\sigma}_c, \end{aligned} \quad (3)$$

where operators L , σ_v , and σ_c operate on the valence electron, valence electron, and conduction electron, respectively. The first three parameters (Δ_i) describe the valence-band crystal-field splitting and the two-component spin-orbit interaction in the valence band, respectively. γ represents the electron-hole exchange interaction, and the remaining quantities are just the four wurtzitic deformation potentials of interest for this paper.⁶ The sp^3 hybridization is the structural basis of group-III semiconductors. This basis causes their conduction band to be determined from s -like antibonding, and their valence bands from p -like bonding orbitals. In the language of elementary group theory,⁷ and including spin, the twofold conduction band transforms like Γ_7 while the symmetry of the sixfold valence band is $\Gamma_9 + \Gamma_7 + \Gamma_7$. We remark that this labeling shows that the irreducible representations of C_{6v} no longer reveal explicitly the notion of spatial parity attached to s and p functions. Description of the exciton *a priori* requires us to construct an appropriate 12-dimensional basis from the valence- and conduction-band Bloch functions. Table I summarizes the basis functions we use to describe the various exciton states. For the purpose of

TABLE I. Basis functions for the $\Gamma=0$ excitons in wurtzite symmetry.

Exciton state	Basis functions
Γ_1	$(p_- \uparrow \beta + p_+ \downarrow) / \sqrt{2}$
Γ'_1	$p_z (\uparrow \alpha + \downarrow \beta) / \sqrt{2}$
Γ_2	$(p_- \uparrow \beta - p_+ \downarrow \alpha) / \sqrt{2}$
Γ'_2	$p_z (\uparrow \alpha - \downarrow \beta) / \sqrt{2}$
Γ_6	$\{p_+ \uparrow \beta, p_- \downarrow \alpha\}$
Γ_5	$\{p_+ \uparrow \alpha, p_- \downarrow \beta\}$
Γ'_5	$\{p_+ \downarrow \beta, p_- \uparrow \alpha\}$
Γ''_5	$\{p_z \downarrow \alpha, p_z \uparrow \beta\}$

the table, from the p -like basis functions, which are noted p_x , p_y , and p_z , we define p_+ and p_- as the following linear combinations: $p_+ = -(p_x + ip_y) / \sqrt{2}$, and $p_- = (p_x - ip_y) / \sqrt{2}$. Spin components of the missing valence electron are \uparrow and \downarrow , and α and β represent the spin components of the conduction electron. The twelvefold exciton transforms according to $3\Gamma_5 + 2\Gamma_1 + 2\Gamma_2 + \Gamma_6$. The construction of such exciton states from Γ_7^c , Γ_9^v , and Γ_7^v states (we add upper scripts to avoid confusion) is sketched in Fig. 1. The twofold Γ_6 exciton is forbidden, as are the two Γ_2 ones. The three Γ_5 exciton are created using an adapted σ -polarized photon (electric field $\perp \langle 0001 \rangle$) and the two Γ_1 excitons are created for π -polarized photons (electric field $\parallel \langle 0001 \rangle$). Due to these symmetry properties, the resolution of the problem requires obtaining eigenstates of low-dimensional matrices. They could eventually be given under the form of analytical equations. We believed it was more convenient for the reader, especially to treat the oscillator strength problem, to use matrix representations. Optically active levels are obtained from two of these matrices: a 3×3 matrix which describes energies of Γ_5 excitons, and a second 2×2 account of the eigenvalues for Γ_1 levels. The last two matrices account for energies of forbidden Γ_2 and Γ_6 excitons.

Eigenenergies of Γ_5 excitons are obtained from resolution of the following matrix:

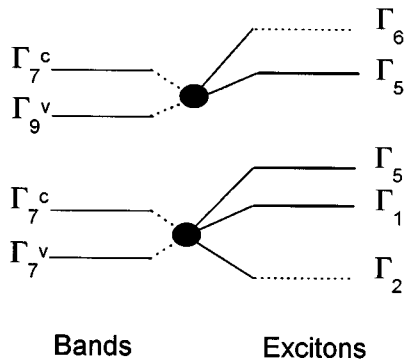


FIG. 1. Schematic construction of excitons (right-hand side) from Bloch states (left-hand side) in α -GaN: Dotted lines correspond to forbidden excitons.

$ \Gamma_5\rangle$	$ \Gamma'_5\rangle$	$ \Gamma''_5\rangle$
$\Delta_1 + \Delta_2 + \delta_1 + \delta_2 - \frac{1}{2}\gamma$	$-\gamma$	0
$-\gamma$	$\Delta_1 - \Delta_2 + \delta_1 + \delta_2 - \frac{1}{2}\gamma$	$\sqrt{2}\Delta_3$
0	$\sqrt{2}\Delta_3$	$\delta_1 + \frac{1}{2}\gamma$

where $\delta_1 = a_z e_{\Gamma_1}^{(1)} + a_x e_{\Gamma_1}^{(2)}$, and $\delta_2 = b_z e_{\Gamma_1}^{(1)} + b_x e_{\Gamma_1}^{(2)}$. The exciton eigenvectors are written $\Psi_{\text{exc}\Gamma_5} = \nu |\Gamma_5\rangle + \omega |\Gamma'_5\rangle + \varphi |\Gamma''_5\rangle$, and their oscillator strengths (σ polarization) are $|\Omega_{\text{exc}\Gamma_5}|^2 = (\nu^2 + \omega^2)/2$.

A similar treatment performed for Γ_1 excitons gives

$ \Gamma_1\rangle$	$ \Gamma'_1\rangle$
$\Delta_1 - \Delta_2 + \delta_1 + \delta_2 + \frac{1}{2}\gamma$	$\sqrt{2}\Delta_3$
$\sqrt{2}\Delta_3$	$\delta_1 - \frac{3}{2}\gamma$

The oscillator strengths of these excitons allowed in π polarization are obtained by the squares of the contribution of $|\Gamma'_1\rangle$ in the two eigenvectors.

For the sake of completeness in this paper, we also address the resolution of forbidden energy levels. The energy of the twofold Γ_6 level is $\Delta_1 + \Delta_2 + \delta_1 + \delta_2 + \frac{1}{2}\gamma$. Concerning Γ_2 states, we have to resolve the following matrix:

$ \Gamma_2\rangle$	$ \Gamma'_2\rangle$
$\Delta_1 - \Delta_2 + \delta_1 + \delta_2 + \frac{1}{2}\gamma$	$\sqrt{2}\Delta_3$
$\sqrt{2}\Delta_3$	$\delta_1 + \frac{1}{2}\gamma$

Making quantitative calculations requires attributing numerical values to all these parameters. From earlier studies of the evolution of A , B , and C lines in σ polarization,⁷ we found that quantities δ_1 and δ_2 are in simple relation: $\delta_1/\delta_2 = -2.136$.^{6,8} Thus a unique parameter— δ_1 , for instance—can be used to describe the stress-induced modification of the internal structure of the excitons in GaN under biaxial stress. We used the following empirical values: $a_x = a_x = -8.16$ eV, $b_z = -2b_x = -1.44$ eV, $\Delta_1 = 10$ meV, $\Delta_2 = 5.1$ meV, and $\Delta_3 = 6.1$ meV.^{6,8}

At the present time, and to the best of our knowledge, γ was neither estimated nor measured in GaN. Its computation is formally extremely tricky, even impossible, without making numerical approximations. From the past 40 years of study of the physics of excitons in crystals, we learned that the influence of the electron-hole exchange interaction on the energy spectrum of the Wannier exciton is large in crystals which exhibit small dielectric constants, or with bands having flat dispersion relations in reciprocal space. As an example of this, let us consider a semiconductor like GaAs, having an exciton Bohr radius of some 150 Å and an exciton binding energy of 4.3 meV. The exchange energy is 0.05 meV.⁹ In another semiconductor like ZnSe with flatter dispersion relations, an exchange energy of ~ 2 meV is estimated,¹⁰ which is only one order of magnitude smaller than the effective Rydberg energy (20 meV). We have calculated γ , extending to the GaN case the two-body calculation developed in Ref. 2 for a few II-VI compounds. The pertinent parameter of the model in Ref. 2 is the product \mathbf{p} of the exciton radius with an *ad hoc* radius k_m of an effective

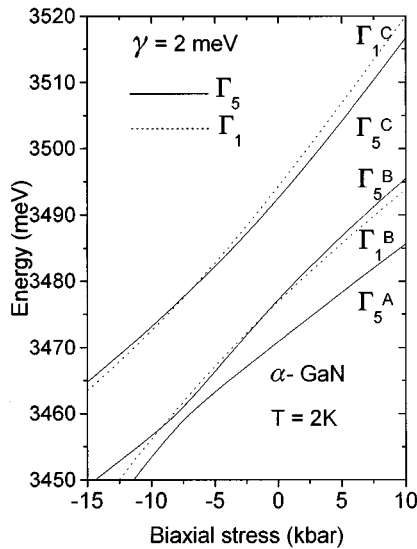


FIG. 2. Evolution of radiative exciton levels as a function of the (0001) biaxial stress. In the picture, positive values of the stress correspond with compression.

Brillouin zone (BZ) hereafter. This k_m is introduced to replace the tedious integration in k space through the whole *actual* BZ by an integration over a spherical space having the volume of this BZ. It has been shown in Ref. 2 that k_m can be connected to the lattice parameter a using a well-adapted scaling argument. For GaN, taking $a=3.19$ Å gives $k_m=0.873$ Å⁻¹. To calculate \mathbf{p} , we need the exciton Bohr radius in GaN. Neither the effective masses nor this quantity are accurately known at present. Recent determination of exciton masses¹¹ suggest $a_B=34\pm 2$ Å. This gives $\mathbf{p}=30\pm 2$, which corresponds to an exchange-influenced $1s$ exciton binding energy of about 85% of R^* ($1/\rho_1^2=0.851\pm 0.006$ in the model of Ref. 2). The exciton exchange energy 2γ is thus estimated to be some 15% of the binding energy. Further, using a Rydberg energy of 26.7 meV the magnitude of the corresponding γ we obtain (~ 2 meV), is far from being negligible, and might have drastic consequence on both (i) the energy spectrum and (ii) the oscillator strength in GaN epilayers under biaxial tension when the crystal-field splitting is offset by the strain field.

Results presented in Fig. 2 display the evolution of Γ_5 (full lines) and Γ_1 (dashed lines) excitons as a function of biaxial stress. In the figure, positive values of the stress correspond to biaxial compression. Using a band-to-band calculation and the parameters we use here, we previously fit the evolution of A , B , and C excitons in epilayers under biaxial compression, i.e., grown on sapphire using different methods.⁶ In these samples, the magnitude of the residual stress is a combined function of both epilayer thickness and growth conditions parameters: V-III molar ratio, interfacial buffer layer, crystalline quality, etc.¹² We note that our calculation predicts that anticrossing of Γ_5 excitons may occur when biaxial tension offsets the crystal-field splitting, e.g., for epilayers deposited on 6H SiC rather than on sapphire. The literature about GaN epilayers grown on 6H-SiC is fairly well documented in terms of a determination of transition energies.¹³⁻¹⁵ In contrast to the case of growth on sapphire, identification of transitions in terms of A , B , and C excitons

is far from being obvious without the help of our calculation.^{6,8,15-18} The optical properties of GaN on SiC are illustrated here using two sets of data we selected from the literature.^{13,14} We note that a sample does exist which corresponds to strain-induced offset of A - B splitting: this degeneracy gives a 3460-meV transition at 2 K.¹³ Now concerning typical mislabeling encountered in the literature, we consider Ref. 14. Using the calculation plotted in Fig. 2, we are now able to interpret transitions detected in reflectometry at 3470–3474 and 3489 meV as the Γ_5 components of A , B , and C excitons. This is at variance with the initial identification of the authors. They interpreted the 3489 line in terms of a B exciton. At this stage, one could—with reason—argue that the deformation potentials are not known accurately, and thus claim that the scale along the x -axis scale of Fig. 2 might slightly deviate linearly from the exact value. This argument does not change the interpretation addressed above, which is also consistent with a more universal plot of the transition energies in a stress-independent diagram, as a function of position of the A line. This approach was earlier used to show a correlation between energies of reflectance structures in GaN grown on sapphire by various methods,^{6,8} and could in addition be extended to 6H-SiC and ZnO.¹⁹ The theory and calculation presented above are restrictive to epilayers grown along the (0001) direction. To the best of our knowledge no quantitative information has been reported, to date, concerning the Γ_5 - Γ_1 splittings in GaN. These measurements require utilizing in-plane propagating photons. Unfortunately, it appears that the homogeneity of strain fields in epilayers prevents this measurement. In addition, thick layers grown by hydride vapor phase epitaxy still display large residual doping and poor structural properties which prevent such fine-structure measurements. An alternative solution could be to grow epilayers on A -plane sapphire substrates. In such a situation, we would face a strong in-plane anisotropy of the optical response. However, the treatment developed above must be slightly modified to account for the interesting symmetry of the problem. This will be subject of a forthcoming publication.

In this section, we compare oscillator strengths for band-to-band and excitonic processes, and we study their evolution with strain. Results are presented in Fig. 3 which concern the σ polarization. The results obtained from the band-to-band calculation are plotted using full lines, while dashed lines correspond to the optical strength for Γ_5 excitons. Band-to-band transitions between Γ_5^c and Γ_5^v keep a constant oscillator strength (0.5 in the notations of the figure) whatever the stress. Band-to-band transitions B and C exchange their oscillator strength when the nature of the residual stress varies from biaxial tension to biaxial compression. We also remark that values expected in a cubic environment ($\frac{1}{6}$ and $\frac{1}{3}$, respectively) are computed when the crystal-field splitting is offset by the biaxial tension. *An exact matching to these values would be obtained if using an isotropic spin-orbit interaction.* The important point we wish to outline here is the remarkable strength of the C band-to-band process or the Γ_5 exciton in σ polarization, for heavily stretched GaN, i.e., for growth on 6H SiC in agreement with reflectance data in Ref. 14. Inclusion of the excitonic effect in σ polarization reinforces the light-crystal coupling for C excitons having Γ_5 symmetry, and simultaneously reduces the oscillator strength

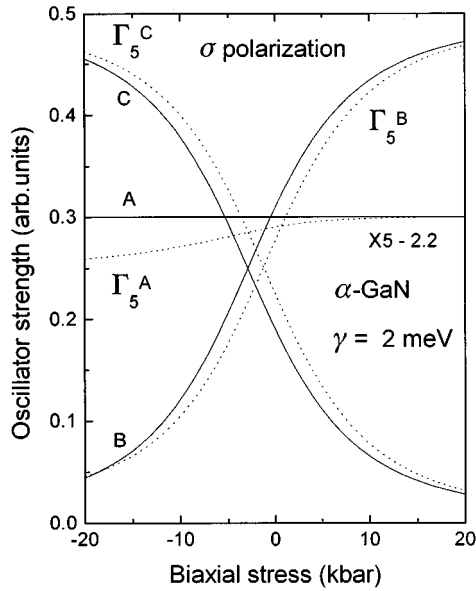


FIG. 3. Oscillator strengths in σ polarization. Full lines: band-to-band calculations. Dotted lines: excitonic calculation. Note that the data for A was magnified five times and then downshifted 2.2 times.

of $\Gamma_5 B$ and $\Gamma_5 A$. Although $\Gamma_5 A$ and $\Gamma_5 B$ states strongly interact for a biaxial tension of about 7.5 kbar, the corresponding exchange of oscillator strengths remains small. Finally, examination of theoretical results for the π polarization is briefly addressed: the two optically active levels also exchange their oscillator strengths as shown in Fig. 4.

Experimental data are in agreement with the predictions of the calculation. Figure 5 displays a series of 2 K reflectivity data taken on a few epilayers grown with different residual strain fields on sapphire substrates, by metal-organic vapor-phase epitaxy. The configuration of the experiment

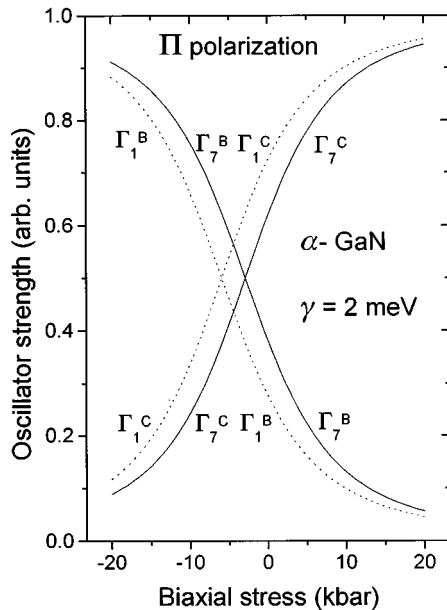


FIG. 4. The analog of Fig. 3, but for π polarization and concerning Γ_1 excitons.

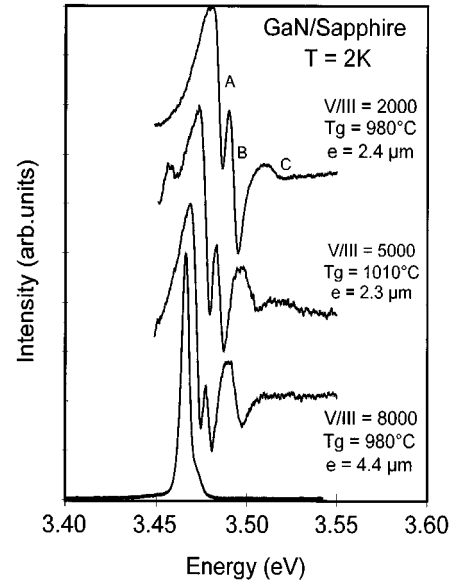


FIG. 5. Plot of typical reflectance features taken in the $\mathbf{E}_{\text{photon}} \perp \langle 0001 \rangle$, $\mathbf{k}_{\text{photon}} \parallel \langle 0001 \rangle$ experimental configuration. Note that the blueshift is accompanied by an increase of the intensity of B and a collapse of line C. The photoluminescence spectrum of the sample giving the reflectance spectrum at the bottom of the picture is also given.

was $\mathbf{E}_{\text{photon}} \perp \langle 0001 \rangle$ and $\mathbf{k}_{\text{photon}} \parallel \langle 0001 \rangle$, so that Γ_5 states could be observed. The first effect we extract from the figure, by comparing features at the top and in the middle of the figure, is that this change of the residual strain is not in simple relation with the thickness of the epilayer. Details about this have been given elsewhere.¹² The absolute value of the light reflectance at energies below the band gap is also sample dependent, due to differences in surface morphology of epilayers grown under various V-III molar ratio, and subsequent macroscopic surface diffusion phenomena. The influence of surface defects on reflectivity was identified many years ago by people working on exciton-polariton phenomena in other bulk crystals, and was demonstrated to alter the background value of the dielectric constant and subsequently the absolute value of the reflectance collected at energies below the band gap due to modification of macroscopic light diffusion phenomena.²⁰ Extensive investigations showed that all these surface topological defects were giving many perturbations, and could be eliminated only after careful polishing and etching of the crystals. Since here we deal with micrometer thin films, any mechanical treatment would be of disastrous influence. As we wish, in this paper, to show strain-related effects on oscillator strengths rather than correlating the absolute reflectance line shape with surface morphology, the experimental data were plotted by scaling them in such a way that amplitudes of A lines are identical from sample to sample. Simultaneously with the blueshift of the three transitions observed when the biaxial compression experienced by the GaN epilayer increases, we observe a collapse of the intensity of C and an increase of the strength of B. This behavior was never analyzed before, to the best of our knowledge, and is in agreement with our theoretical predictions in Fig. 3.

In conclusion, we tried to outline the fact that GaN epi-

layers grown with wurtzite symmetry may display a large variety of optical properties depending on the residual stress in the epilayer. We found, in particular, that the strength of optical transitions is extremely sensitive to the residual strain field. This has a significant influence on the line shape of optical features which are detected in adapted experiments like reflectivity, due to the small value of the spin-orbit interaction. The calculation was done in the context of two

models, one of them including the excitonic exchange interaction which we estimated in GaN.

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