The influence of phonons on the optical properties of GaN

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We comprehensively examine the importance of phonons on the optical properties of GaN. Using Raman and photoluminescence (PL) spectroscopies, the energies and linewidths of optic phonons, excitons, and discrete phonon sidebands (PSBs) are studied between 20 and 325 K. The temperature dependence of the A_1 (LO) phonon energy and linewidth are described by a combined two- and three-phonon decay process. The narrow E_2^2 phonon decays by the three-phonon emission process. Three band-edge excitons are observed in PL with linewidths between 2.8 and 5.3 meV at temperature 22 K. The energy gap shrinkage and exciton linewidths are completely interpreted based on electron-phonon interactions. The shift, broadening, and asymmetry of the PSBs are explained by incorporating the decay mechanism of A_1 (LO) phonon and the exciton broadening from electron-phonon interactions. © 2006 American Institute of Physics.

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I. INTRODUCTION

The lifetimes of intrinsic phonons in high-quality crystalline materials are determined by phonon decay processes.^{1,2} The decay mechanisms are governed by energy and wave vector conservation, and specific decay channels are important when those vibrations created have high density of states. Phonon decay is important to understand in materials when devices based on them exhibit self heating, such as GaN-based bipolar and field-effect transistors.³ Optical properties of semiconductors are useful for understanding the electronic band structure. This is particularly important for states near the valence and conduction edges which are the most relevant to light emitting devices and to carrier transport. The band structure and behavior of carriers in such materials are strongly influenced by lattice vibrations. For direct optical transitions the zone-center phonons are most relevant. In the III-nitride semiconductors, e.g., GaN, bulk crystals are not widely available. Consequently, studies of these semiconductors have primarily relied on heteroepitaxy using foreign substrates.^{4,5} The resultant epitaxial crystals are generally abundant in dislocations. This situation is in stark contrast to other semiconductors for which high-quality bulk crystals are routinely available, and hinders study of intrinsic GaN properties.

Recent advances in growing epitaxial GaN, with relatively low defect density,⁶ present the opportunity to study fundamental optical processes across a broad temperature range. Narrow phonon lines observed in high-quality crystals are likewise necessary to examine the decay properties of the zone-center phonons of GaN. The decay is examined through temperature dependence of the Raman-active $\mathbf{k}=0$ phonon energy and linewidth. GaN crystals with low dislocation den-

sity enable examination of the band-edge excitons, through photoluminescence (PL) spectroscopy, and the properties of discrete A_1 (LO) phonon sidebands (PSBs).

We present a study of phonon decay and the influence of phonons on excitons and the band gap of GaN. Using Raman scattering, we study the decay properties of E_2^2 and longitudinal-optic A_1 (LO) symmetry phonons. We also examine the near-band gap PL to study the temperature dependence of the GaN energy gap, via the exciton emission, and their A_1 (LO) PSBs. The band gap shrinkage, the exciton linewidth, and the PSBs are all strongly dependent on the vibrational properties. The results are remarkably consistent, providing a comprehensive examination of the impact phonons have on the optical properties of GaN.

II. EXPERIMENTAL DETAILS

The nominally undoped GaN layers, with layer thickness ~ 80 μm , were grown using hydride vapor phase epitaxy on 2-in. sapphire substrates.⁷ The layers had smooth surface morphology with root-mean-square (rms) roughness below 0.5 nm as measured by atomic force microscopy using 5×5 μm^2 scans. No cracks were observed. The concentration of electrically active (noncompensated) donors was in the low 10^{16} cm^{-3} range from capacitance-voltage measurements. X-ray diffraction reveals layers to have low dislocation density, $<10^8$ cm^{-2} , and to be strain relaxed.⁸

Micro-Raman spectra were measured in a backscattering geometry using excitation wavelength 488.0 nm.⁹ Micro-PL spectra were obtained with 351.0 nm excitation and dispersed using a 0.78 m double monochromator. Linewidths have been corrected for the instrumental bandpass. Excitation power was kept well below 10^4 W/cm² to minimize local heating. Temperature was controlled between ~ 20 and 325 K using a closed-cycle cryostat. In this article we use the conventional notation of ω for phonon energy in cm^{-1} and ϵ for energy in electron-volts or milli-electron-volts.

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FIG. 1. Raman spectrum of GaN epilayer at 20 K.

III. E_2^2 AND A_1 (LO) PHONONS

The Raman spectrum for the GaN sample at $T=20$ K is shown in Fig. 1. The Raman shift and linewidth (Γ , full width at half maximum) are determined by fitting Lorentzian functions to the data. Two features at 569 and 737 cm^{-1} correspond to the E_2^2 and A_1 (LO) phonons.¹⁰ The E_2^2 and A_1 (LO) intensities are as expected for GaN with low free-carrier concentration.¹¹ The linewidths of E_2^2 and A_1 (LO) at $T=20$ K compare well with previously published reports^{12–15} indicating high crystalline quality material.

In Fig. 2 we show the temperature dependences of the phonon energies and linewidths. The Raman bands redshift with temperature and broaden, although the changes are stronger for the A_1 (LO) line than for E_2^2 . The temperature dependences of $\omega(T)$ can be described by the crystal thermal expansion, anharmonic interactions, and lattice and thermal mismatches between the substrate and the epitaxial layer

$$\omega(T) = \omega_0 - \Delta_1(T) - \Delta_2(T), \quad (1)$$

where ω_0 is the harmonic frequency of the phonon mode, $\Delta_1(T)$ is the thermal expansion contribution to the frequency, and $\Delta_2(T)$ is the anharmonic interaction term. The thermal strain is negligible [<0.1 cm^{-1} shift for the A_1 (LO) phonon] across the temperature range studied.¹⁶ The thermal expansion contribution is given by $\Delta_1(T) = \omega_0 \gamma \int_0^T [\alpha_c(T') + 2\alpha_a(T')] dT'$, where γ is the Gruneisen parameter, and α_c and α_a are the temperature-dependent linear thermal expansion coefficients parallel and perpendicular to the hexagonal c axis, respectively.¹⁷ The anharmonic shift $\Delta_2(T)$ is¹⁶

$$\begin{aligned} \Delta_2(T) = & A[1 + n(\omega_1, T) + n(\omega_2, T)] \\ & + B[1 + n(\omega_3, T) + n(\omega_4, T) + n(\omega_5, T) \\ & + n^2(\omega_3, T) + n^2(\omega_4, T) + n^2(\omega_5, T)], \end{aligned} \quad (2)$$

where $n(\omega, T) = [\exp(\hbar c \omega / k_B T) - 1]^{-1}$ is the Bose function at energy $\hbar c \omega$. The first term in Eq. (2) corresponds to the two-phonon decay process, where ω_1 and ω_2 are frequencies of the phonons created by the decay. The second term in Eq. (2) corresponds to the three-phonon decay process with en-

FIG. 2. Dependence of Raman-active phonon energies and linewidths on temperature: (a) and (b) are for A_1 (LO), (c) and (d) are for E_2^2 . Curves are fits to data. Different contributions to the phonon shifts are plotted separately in (a) and (c), respectively.

ergies ω_3 , ω_4 , and ω_5 . The created/annihilated phonon energies must satisfy conservation of energy. For the zone-center Raman-active phonons, crystal momentum conservation requires the sum of the wave vectors of the created phonons in the decay to be zero. Anharmonic constants A and B identify the relative probabilities of the decay processes.

The Raman linewidth can be described in terms of anharmonic interactions¹⁸

$$\Gamma_P(T) = \Gamma_0 + \Delta'_2(T), \quad (3)$$

where $\Delta'_2(T)$ is identical in form to $\Delta_2(T)$ with A (B) replaced by C (D), and the ω_i energies are the same as what we obtain from fitting Eq. (2) to the phonon energy data. With

TABLE I. Fitting results for Raman phonons and band gap related excitons. All $T=0$ K values are from fits to the temperature dependences.

Excitation	Energy	Linewidth
E_2^2	$\omega_0=569.1 \text{ cm}^{-1}$ $A=0, B=0.09 \text{ cm}^{-1}$ $\omega_3=\omega_4=\omega_5=190 \text{ cm}^{-1}$	$\Gamma_0=1.4 \text{ cm}^{-1}$ $C=0, D=0.10$
A_1 (LO)	$\omega_0=738.7 \text{ cm}^{-1}$ $A=2.2 \text{ cm}^{-1}, B=0.09 \text{ cm}^{-1}$ $\omega_1=\omega_3=559 \text{ cm}^{-1}, \omega_2=180 \text{ cm}^{-1}$ $\omega_4=\omega_5=90 \text{ cm}^{-1}$	$\Gamma_0=1.4 \text{ cm}^{-1}$ $C=1.9 \text{ cm}^{-1}, D=0.2 \text{ cm}^{-1}$
X_A	$E(0)=3.473 \text{ eV}$ $\epsilon_{ac}=22.5 \text{ meV}=181 \text{ cm}^{-1}$ $\epsilon_{opt}=69.3 \text{ meV}=559 \text{ cm}^{-1}$ $W_{ac}=0.49, W_{opt}=0.51$	$\Gamma(0)=5.8 \text{ meV}=47 \text{ cm}^{-1}$ $\Gamma_{ac}=50 \text{ meV}=400 \text{ cm}^{-1}$ $\Gamma_{opt}=110 \text{ meV}=890 \text{ cm}^{-1}$
X_B	$E(0)=3.480 \text{ eV}$	
(D^0, X)	$E(0)=3.467 \text{ eV}$	$\Gamma(0)=3.7 \text{ meV}=30 \text{ cm}^{-1}$

these constraints, the number of fitting parameters inherent in the phonon-decay model is held to a minimum. Γ_0 is related to the phonon lifetime and is affected by inherent disorder or impurity scattering in high-quality material. If we attribute the A_1 (LO) phonon linewidth to homogeneous broadening only, the lifetime obtained is ~ 1.6 ps which compares well with measurements using time-resolved spectroscopy.¹⁹

We use Eqs. (1) and (3) to fit the dependence of our $\omega(T)$ and $\omega(T)$ data, as shown in Figs. 2(a) and 2(b). Fit results are included in each panel and parameters are summarized in Table I. For the Raman energy we show the individual contributions of the thermal expansion, Eq. (2) shifted by ω_0 , and two-phonon decay process. These terms produce comparable contributions to the overall shift. The peak energy and linewidth results consistently show that two-phonon decay plays a larger role than the three-phonon mechanism in the A_1 (LO) phonon temperature dependence ($A \gg B$ and $C \gg D$). The two-phonon energies 559 and 180 cm^{-1} are consistent with $\text{LO} \rightarrow \text{TO} + \text{LA(TA)}$ decay.^{20,21} This decay is referred to as the Ridley process. The three-phonon decay energies 560 , 90 , and 90 cm^{-1} are consistent with the $\text{LO} \rightarrow \text{TO} + \text{LA(TA)} + \text{LA(TA)}$ three-phonon decay in Refs. 2 and 21. The two-phonon decay is dominant in the temperature dependence of the A_1 (LO) phonon behavior.

In contrast to the A_1 (LO) phonon, results in Fig. 2(c) show the E_2^2 energy to depend weakly on temperature, red-shifting by only $\sim 1.5 \text{ cm}^{-1}$ across the temperature range studied. This is attributed to the absence of two-phonon decay channels satisfying both conservation of energy and wave vector criteria and exhibiting appreciable density of states. Thus, $A=0$ in Eq. (2) for the E_2^2 phonon. In Fig. 2(c) we plot the individual contributions from the dominant thermal expansion and the three-phonon decay process to the overall Raman shift for E_2^2 mode.^{14,22} The decay creates three acoustic phonons at $\sim 190 \text{ cm}^{-1}$.¹³ The linewidth in Fig. 2(d) also varies weakly with temperature, consistent with three-phonon decay. The dependence is fitted well with $C=0$ and $D \neq 0$ and using the same decay energies. Our results show

FIG. 3. Photoluminescence spectrum of GaN epilayer at 22 K. Donor-bound exciton line labeled (D^0, X) , X_A and X_B are free excitons. PSB1 and PSB2 denote A_1 (LO)-phonon sidebands.

the contribution from three-phonon decay is very small throughout this temperature range. This is in contrast to previous Raman work, which concluded that the thermal expansion and three-phonon process made comparable contributions.^{14,22} It is important to note that we analyze our phonon energy and linewidth data and that we use the temperature dependent expansion coefficients, which vary significantly over the range of our experiments.¹⁷

IV. BAND-EDGE EXCITONS

The phonon properties are critical in understanding the temperature dependence of the energy gap. In Fig. 3 the low-temperature PL spectrum exhibits three exciton bands at 3.468 , 3.475 , and 3.482 eV . Our exciton lines agree with those of unstrained materials, as expected from our x-ray measurements, identifying these features as the shallow neutral donor-bound (D^0, X) and free excitons (X_A and X_B).²³ Each line is narrow at this temperature, 2.8 , 5.3 , and 4.7 meV , respectively. We see two A_1 (LO) PSBs, originating from the X_B exciton, with the characteristic asymmetry attributed to interactions between discrete and continuum states.²⁴

In Fig. 4(a) we summarize the exciton peak energies as a function of temperature. The characteristic redshift with increasing temperature is attributed to band gap reduction from lattice expansion and to electron-phonon interactions.²⁵ Both factors are closely associated with phonon populations. We apply the approach of Pässler,²⁶ which incorporates both self-energy Fan²⁷ and Debye-Waller²⁸ factors. The resulting expression for the temperature shift of the band gap is $\Delta E_{e-p}(T) \rightarrow -(\delta/k_B) \sum_{i=1}^N \epsilon_i W_i n(\epsilon_i, T)$, where δ is the high-temperature linear slope of the band gap, and the weighting factor is normalized $\sum_{i=1}^N W_i = 1$. We adopt a simple two-oscillator ($N=2$) model, corresponding to acoustic and optic phonon branches, and consider the contribution from thermal expansion. The temperature dependence is thus

FIG. 4. Temperature dependences of exciton energies (a) and linewidths (b). Temperature dependences of LO-phonon PSB energies (c) and linewidths (d). The second PSB energy ω_{PSB2} is scaled by a factor of 2 so that it can be directly compared with ω_{PSB1} . Curves are theoretical fits to the data.

$$E(T) = E(0) - \frac{\delta}{k_B} [\varepsilon_{ac} W_{ac} n(\varepsilon_{ac}, T) + \varepsilon_{opt} W_{opt} n(\varepsilon_{opt}, T)] - \int_0^T [\xi_c \alpha_c(T') + 2\xi_a \alpha_a(T')] dT', \quad (4)$$

where the last term accounts for thermal expansion.²⁹ Quantities ξ_a and ξ_c are deformation potentials in the a and c crystal directions, respectively.³⁰

The temperature dependence of the band gap and the fit to data using Eq. (4) are shown in Fig. 4(a). The four independent fitting parameters in Eq. (4) are also related to the

exciton broadening discussed below. Table I summarizes the fit results. We break out the various contributions to the shift from the electron-phonon interaction $\Delta E_{e-p}(T)$. The majority of the temperature dependence stems from the electron-phonon contribution. Our results show the two-phonon Pässler approach accurately describes the temperature dependence of the GaN band gap. The phonon energies in the model are ~ 23 meV = 186 cm⁻¹ and ~ 69 meV = 557 cm⁻¹, respectively, corresponding to weighted averages of the acoustic and optic density of states, respectively.²⁶

The temperature dependence of the exciton linewidths, Fig. 4(b), is attributed to phonon-induced band broadening³¹

$$\Gamma(T) = \Gamma(0) + W_{ac} \Gamma_{ac} n(\varepsilon_{ac}, T) + W_{opt} \Gamma_{opt} n(\varepsilon_{opt}, T), \quad (5)$$

where $\Gamma(0)$ is the inhomogeneous linewidth due to intrinsic lifetime, electron-electron interactions, defects, and strain. The second term in Eq. (5) arises from acoustic phonons and the third term is from optic phonons. In the intermediate temperature range, $\varepsilon_{ac} < k_B T < \varepsilon_{opt}$ we recover the less general approximation $n(\varepsilon_{ac}, T) \propto T$.³¹ W_{ac} and W_{opt} are fixed at the values obtained for the energy gap dependence, Eq. (4), and the phonon energies ε_{ac} and ε_{opt} are likewise the values obtained from that analysis. Γ_{ac} and Γ_{opt} are the electron-phonon coupling constants.²⁹ Treating $\Gamma(0)$, Γ_{ac} , and Γ_{opt} as fitting parameters, we obtain $\Gamma(0) \sim 3.7$ and 5.8 meV for the (D^0, X) and X_A excitons, respectively. The intensity of the X_B band decreases quickly with higher temperature. Below $T = 200$ K its temperature dependence follows the same trend as the other exciton bands. We thus rely on (D^0, X) and X_A in obtaining fitting parameters. We obtain values for $\Gamma_{ac} \sim 50$ meV and $\Gamma_{opt} \sim 110$ meV. Direct comparison may be made with other studies, although composite quantities $W_{ac} \Gamma_{ac}$ and $W_{opt} \Gamma_{opt}$ are generally published as a single fitting parameter. The results are in good agreement with theoretical³² values for the optic phonons, 60.2 meV to compare with our $W_{opt} \Gamma_{opt} = 55$ meV. It is substantially lower than the previous experimental value $W_{opt} \Gamma_{opt} = 100$ meV,¹¹ which we attribute to the different methods used in describing the acoustic contribution to the linewidth and to the low dislocation density of our current materials.

V. PHONON SIDEBANDS

We now turn our attention to the temperature dependence of the A_1 (LO) PSBs in Fig. 4. Summarized in Figs. 4(c) and 4(d) are the PSB energies and linewidths, extending the range previously studied to below 50 K.³³ We graph energy $\omega_{PSB2}/2$ for direct comparison with ω_{PSB1} . Γ_{PSB1} and Γ_{PSB2} are not rescaled. PSB properties have been studied theoretically²⁴ and experimentally,^{33,34} with interesting results. The shift in ω_{PSB1} is considerably larger than for $\omega_{PSB2}/2$. Further, Γ_{PSB1} is found to be significantly narrower than Γ_{PSB2} . Prior work focused on the importance of the exciton-phonon density of states in the broadening and shift. The transition probability is $P_m(\omega) \propto \chi_m^{(5/2)-m} \exp(-\chi_m/k_B T)$, where $\chi_m = \hbar\omega + m\hbar\omega_{LO} - E_{exc}$, $m(=1, 2)$ is the PSB index, and E_{exc} is the exciton energy.²⁴ Physically, the prefactor originates from the density of states and the Boltzmann factor arises from the Fermi tail describing the thermal popula-

tion of the exciton. This theory may be used to describe the PSB shift and linewidth as a function of temperature,³³ but it does not account for the phonon broadening expected with increasing temperature. Including phonon broadening, we expect the energy to depend on temperature according to

$$\omega_{\text{PSB}m}(T) = m\omega_0 - \left(\frac{5}{2} - m\right)k_B T - m\Delta_2(T), \quad (6)$$

where ω_0 and $\Delta_2(T)$ are for the A_1 (LO) phonon, Eqs. (1) and (2), taken directly from the Raman analysis. The linear term in T arises from the shift in $P_m(\omega)$. Since ω_{PSB} is relative to the exciton, the band gap shift of Eq. (4) is absent. Agreement between the data and Eq. (6) is excellent for both sidebands. The PSB shifts are dominated by terms linear in T while the phonon decay plays a small role over this temperature range. The broadening exhibits significant curvature in Fig. 4(d), suggesting that the phonon-related processes are important in the dependence. The PSB widths also have a term linear in temperature. Using the Segall–Mahan theory and including the exciton and phonon broadening factors, the sideband linewidths vary according to

$$\Gamma_{\text{PSB}m}(T) = 2\sqrt{\left(\frac{5}{2} - m\right)k_B T + \Gamma(T) + \Gamma_p(T)}, \quad (7)$$

where $\Gamma(T)$ is from Eq. (5) and $\Gamma_p(T)$ is from Eq. (3). The latter term is negligible since the A_1 (LO) band is very narrow compared to the other terms in Eq. (7). Fit results are included in Fig. 4(d). The $\Gamma(0)$ values of 6.0 and 4.5 meV for PSB₁ and PSB₂ are close to the exciton linewidths at low temperature. The phonon energies for $\Gamma(T)$ are the same as what we obtained for the excitons in Table I. The exciton broadening parameters from the X_A line are used. However, the temperature dependent term in Eq. (5) is ten times larger for the PSBs than for the excitons. This may be associated with the rapid rise seen in the X_B linewidth below 100 K, Fig. 4(b). Physically, this broadening may stem from the elevated electron-phonon interactions, attributed to quantum interference between intermediate states of excitons via the LO phonons. In GaN, the participating states are the separate exciton levels and these effects give rise to the asymmetric Fano line shape of the phonon sidebands.^{24,33,34}

VI. SUMMARY

In summary, we have investigated the influence of phonons on the temperature dependence of optical transitions in GaN. By comprehensively studying the phonon properties and the band gap-related emission we examine the relationship between vibrational and electronic properties. Both phonons and excitons are narrow, indicative of high-quality GaN. The observed band gap dependence is fully described based on the Fan and Debye–Waller factors using the approach of Pässler.²⁶ Exciton broadening is also found to depend solely on the phonon band structure which is consistent with results of the energy gap dependence. The temperature dependence of the A_1 (LO) PSBs are described using the Segall–Mahan theory,²⁴ with the phonons taken into account. We find the A_1 (LO) phonon properties play a minor role in the PSB energies, but play a strong role in the line broaden-

ing. Our results are highly relevant to understanding the relationship between electronic bands and phonons in related semiconductors, particularly AlN and InN which have the wurtzite crystal structure but exhibit widely varying energy band structures.

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