

Comparative study on the broadening of exciton luminescence linewidth due to phonon in zinc-blende and wurtzite GaN epilayers

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Linewidth broadening of exciton luminescence in wurtzite and zinc-blende GaN epilayers was investigated as a function of temperature with photoluminescence. A widely accepted theoretical model was used to fit the experimental data, so that the coupling parameters between exciton and acoustic and longitudinal optical phonons were obtained for both structures. It was found that the coupling constants of both exciton–acoustic optical phonon coupling and exciton–longitudinal optical phonon coupling for zinc-blende GaN are almost twice as much as the corresponding values of wurtzite GaN. These results show that the relatively strong exciton–phonon scattering seems to be characteristic to zinc-blende GaN film. © 2002 American Institute of Physics.
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It is known that GaN exists in two different crystalline structures under usual conditions. The equilibrium phase is wurtzite GaN (α -GaN), while the metastable zinc-blende GaN (β -GaN) can be epitaxially grown. In the past a few years, the structural, optical, and electrical properties of α -GaN as well as α -GaN-based devices have been extensively studied.^{1–3} However, many outstanding issues remain unsolved. Compared with α -GaN, fewer studies⁴ have been devoted to β -GaN. This situation will change, as rapidly increasing attention has been paid to the system in recent years.^{5–9} Yang *et al.*¹⁰ and As *et al.*¹¹ recently demonstrated electroluminescence from β -GaN p – n junctions, which represents the starting point of β -GaN based optoelectronic devices. Theoretical work has predicted that β -GaN has different properties from α -GaN, for example, smaller band-gap^{12,13} and easier p -type doping.¹⁴ Some of these have already been verified experimentally.

In this letter, a comparative study on exciton scattering by phonons in β -GaN and α -GaN films is reported. The temperature dependence of the linewidth of exciton lines in the two GaN films is fitted with a theoretical model. It was found that the coupling strength of the excitons with both the acoustic phonons and the LO phonons in α -GaN is much smaller than the corresponding ones in β -GaN. The relatively strong exciton–phonon coupling may thus be characteristic of β -GaN.

The β -GaN films were grown on Si-doped GaAs (001) substrates by metalorganic vapor phase deposition using ammonia and trimethylgallium as N and Ga precursors, respectively. H₂ was used as the carrier gas during deposition. A nucleation GaN layer was first deposited at 550 °C for 10 min. It was followed by a 2- μ m-thick β -GaN layer at 820 °C. The α -GaN epilayers were directly grown on 6H-

SiC (0001) substrates at 650 °C by plasma-assisted molecular beam epitaxy (MBE).¹⁵ The thickness of the α -GaN epilayer was about 1.8 μ m. A micro-Raman system was employed to characterize the phonon structures of the two GaN films at room temperature. The excitation laser source in the Raman scattering was the 514.5 nm line of an Ar⁺ laser. The incident laser beam is along the growth direction of the films and the back-scattering geometry was taken. The photoluminescence (PL) setup has been described previously (see Ref. 22). In the PL measurements, the same measurement conditions were applied to both samples.

Figure 1 shows the measured PL spectra from the β -GaN and the α -GaN samples at different temperatures. At low temperature, the peaks at 3.269 and 3.151 eV from β -GaN have been identified by many authors^{4–6,9} as the shallow donor-bound exciton and the donor–acceptor pair transition lines. Our varying excitation power experiment also presents a clear spectral evidence that the peak at about 3.151 eV is the donor–acceptor pair transition. The linewidth of the donor-bound exciton line of β -GaN at 3.5 K is 14.6 meV, which is comparable to the best values reported so far.⁵ The linewidth of the dominant peak broadens to 54.9 meV at 300 K. To the best of our knowledge, this is the narrowest PL linewidth observed at room temperature for β -GaN.^{4,5} Moreover, the successful demonstration of electroluminescence from β -GaN p – n junction¹⁰ also verified the high quality of the sample. In general, the shallow donor-bound exciton transition dominates the low temperature PL spectrum of α -GaN. From Fig. 1, it can be seen that the linewidth of the dominant bound exciton line is 12.8 meV at 3.5 K, which is close to that of the β -GaN. The linewidth of the exciton line increases from 12.8 to 32.2 meV when the temperature increases from 3.5 to 300 K.

The temperature dependence of the linewidth of the lowest 1S exciton transition in semiconductors can be given by¹⁶

$$\Gamma(T) = \Gamma_0 + \sigma T + \gamma N_{LO}(T). \quad (1)$$

The first term on the right-hand side is a constant, standing

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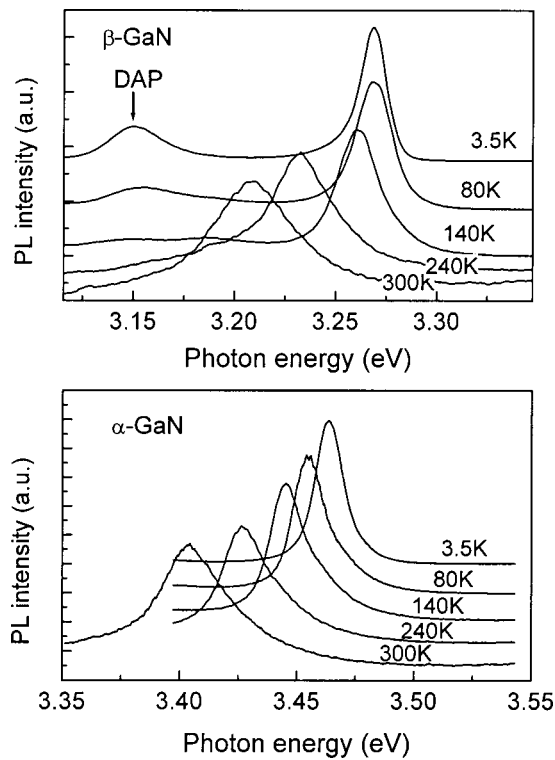


FIG. 1. Measured PL spectra of the zinc-blende GaN and the wurtzite GaN heteroepilayers at different temperatures. For clarity, all the spectra were normalized and were shifted vertically except one at 300 K.

for the impurity and imperfection scattering. The second term arises from the scattering of the excitons by acoustic phonons, exhibiting the linear temperature dependence. σ is the exciton-acoustic phonon coupling strength. The last term is due to the interaction of the excitons with the LO phonons and is proportional to the Bose function $N_{LO}(T) = [\exp(\hbar\omega_{LO}/k_B T) - 1]^{-1}$ for LO-phonon occupation, where $\hbar\omega_{LO}$ is the LO phonon energy and γ represents the exciton-LO phonon coupling strength. The above theoretical model has been frequently employed to study the thermal broadening of exciton luminescence in various semiconductors. For α -GaN, several groups¹⁷⁻¹⁹ fitted the experimental linewidth using Eq. (1).

Figure 2 shows the linewidth of the exciton luminescence in α -GaN and β -GaN as a function of temperature. The solid lines represent the least-squares fitting of the data considering temperature-independent inhomogeneous broadening, acoustic phonon scattering, and LO phonon scattering. As a highly polar semiconductor, GaN possesses much stronger exciton-LO phonon scattering than other III-V semiconductors, such as GaAs. In order to accurately determine the characteristic energy of LO phonons in the β -GaN and the α -GaN films, Raman scattering spectra of both samples were taken at room temperature, as shown in Fig. 3. The measured phonon energies of the α -GaN and the β -GaN samples are in good agreement with the values reported in the literature.^{7,8,20,21}

From Fig. 3, it can be seen that a low content of α -GaN exists in the β -GaN host because of the resolved E_2 phonon mode, which is a characteristic mode of α -GaN. It should be pointed out that the E_2 phonon mode is not always observable when we change the excitation spot. This implies that the low content of the α -GaN phase in the β -GaN film is not

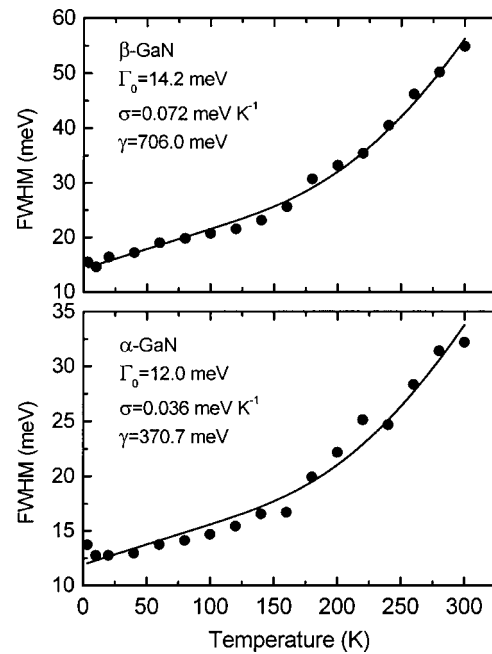


FIG. 2. Temperature dependence of the linewidth for two GaN epilayers. Solid lines are the fitting results with Eq. (1).

uniformly distributed. We took the measured energies of the LO phonons when fitting the linewidth in Fig. 2. It is found that the result is excellent with the following parameters: $\Gamma_0 = 14.2$ meV, $\sigma = 0.072$ meV K⁻¹, and $\gamma = 706.0$ meV for β -GaN, and $\Gamma_0 = 12.0$ meV, $\sigma = 0.036$ meV K⁻¹, and $\gamma = 307.7$ meV for α -GaN. It is noted that the σ and γ values obtained here for α -GaN are consistent with those obtained by Buyanova *et al.*¹⁷ Obviously, the σ and γ values for β -GaN are much greater than the corresponding ones of α -GaN, although their Γ_0 values are comparable. This suggests that there exists a stronger exciton-phonon scattering in β -GaN. Brandt and his co-workers⁴ also observed a large thermal broadening of the near-band edge transition in β -GaN. Furthermore, they pointed out that this was a characteristic property of β -GaN.

According to Rudin *et al.*'s theoretical calculations,¹⁶ the contribution of exciton-LO phonon scattering to the linewidth is mainly from the Fröhlich interaction. In general, the larger LO phonon energy favors stronger Fröhlich interaction

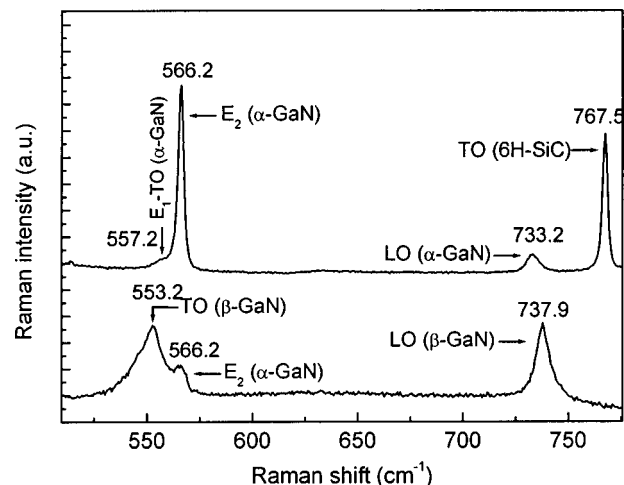


FIG. 3. Raman scattering spectra for two GaN epilayers at room temperature.

and thus a larger coupling strength γ . From Fig. 3 we know that there is a slightly larger LO phonon energy for β -GaN than for α -GaN. This small energy difference is perhaps particularly important for polar semiconductors like GaN with very intense exciton-LO phonon Fröhlich scattering. Another important factor relating to the thermal broadening of the exciton lines is the binding energy of excitons.¹⁶ It is very likely for β -GaN^{22,23} to have smaller binding energy of free excitons than α -GaN.^{24,25} At higher temperatures, the free exciton recombination generally dominates in the PL spectra of GaN. This can be judged from the characteristic asymmetric lineshape of the free exciton emission.²⁶ Note that the lineshape of the dominant emission line from the β -GaN film changes from asymmetric to symmetric when temperature is above 240 K. But for the α -GaN sample, the emission line keeps its characteristic asymmetric lineshape from 80 up to 300 K. This fact also indicates that β -GaN has relatively smaller binding energy for free excitons. It can be expected that β -GaN will have relatively broad emission line at higher temperatures. Therefore, both factors, the relatively larger LO phonon energy and relatively smaller free-exciton binding energy, will cause significant thermal broadening of the emission line in β -GaN, particularly, at higher temperatures.

At low and medium temperatures, the contribution of acoustic phonon scattering to the broadening of exciton line dominates. The exciton-acoustic phonon interaction takes place via the deformation potential and the piezoelectric coupling. The contribution of deformation potential scattering to the linewidth is approximately equal for wurtzite and zinc-blende GaN films due to the nearly equal deformation potentials of GaN with the two crystalline structures.²⁷ However, the contribution of piezoelectric interaction is different.¹⁶ Thus, we believe that the difference between the exciton-acoustic phonon coupling constants of β -GaN and α -GaN is primarily due to different piezoelectric interactions for the two structures.

In order to further confirm the relatively stronger coupling of exciton with LO and acoustic phonons in β -GaN, we plotted the temperature dependence of the peak positions of the dominant emission lines for two GaN in Fig. 4. It is seen that the peak positions have almost same temperature dependence between β -GaN and α -GaN, but there is a shift of 195 meV in energy between the two. Note that the α -GaN and β -GaN films investigated here were grown on different substrates. The same temperature dependence of the emission photon energies between the two GaN films provides an additional evidence that the relatively larger exciton-phonon scattering is an intrinsic property of the β -GaN epilayers.

In conclusion, the temperature dependence of the dominant emission lines from the GaN epilayers in wurtzite and zinc-blende structures has been compared. A widely accepted theoretical model was employed to fit the experimental data. The relative stronger scattering of exciton with both LO and acoustic phonons was found in the β -GaN epilayer. Analysis shows that it is a characteristic property of β -GaN.

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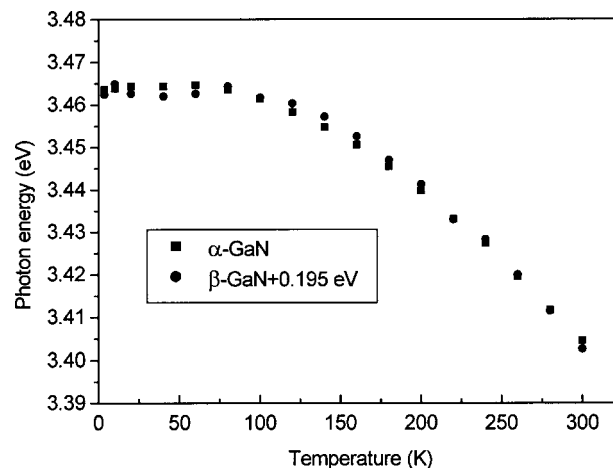


FIG. 4. Temperature dependence of the peak positions of the dominant emission for two GaN epilayers.

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