Exciton-phonon interactions, exciton binding energy, and their importance in the realization of room-temperature semiconductor lasers based on GaN

Annamraju Kasi Viswanath
National Creative Research Initiatives Center for Characterization of Ultrafast Optical Properties, Korea Research Institute of Standards and Science, Yusong, Taejon 305-600, Korea

Joo In Lee
Spectroscopy Laboratory, Korea Research Institute of Standards and Science, Yusong, Taejon 305-600, Korea

Dongho Kim
National Creative Research Initiatives Center for Characterization of Ultrafast Optical Properties, Korea Research Institute of Standards and Science, Yusong, Taejon 305-600, Korea

C. R. Lee and J. Y. Leem
Epitaxial Semiconductor Group, Korea Research Institute of Standards and Science, Yusong, Taejon 305-600, Korea
(Received 27 April 1998; revised manuscript received 25 August 1998)

Temperature dependence of the linewidths of free-exciton *A* and *B* transitions was investigated. Experimental linewidths were fitted to a theoretical model considering various interactions of excitons with phonons in addition to inhomogeneous broadening. It was shown that acoustic phonon scattering must also be considered to explain the emission linewidth broadening, in contrast to a recent report on luminescence linewidths in GaN. These exciton–acoustic-phonon interactions also explain the fast energy relaxation of free excitons to the bottom of the exciton band, which leads to generally observed short free-exciton lifetimes in GaN. The exciton–longitudinal-optical (LO) -phonon coupling constant was found to be extremely large. This was explained as being due to the Fröhlich interaction and the polar nature of GaN. The binding energy of both *A* and *B* excitons was found to be 26 meV. The relevance of exciton-phonon interactions and the binding energy of free excitons in achieving room-temperature exciton-based semiconductor lasers was discussed. Though exciton–LO-phonon interaction was very strong in GaN, it was still possible to observe room-temperature excitons since the exciton binding energy is very large. [S0163-1829(98)02648-4]

When semiconductors are optically excited, several interactions take place among excitons, free carriers, lattice phonons, impurities, and defects. Such interactions have been studied in bulk and quantum-confined semiconductors by absorption, luminescence, coherent spectroscopies in time, and frequency domain. Particularly the coupling between excitons and phonons is an important parameter in determining the electronic and optical properties of semiconductors. Recently, GaN and other III-nitrides have received considerable attention due to the possibility of fabricating blue and UV light sources and detectors.[1] The applicability of these materials from the point of view of excitonic devices depends upon the strength of the room-temperature exciton resonances. Therefore, the width of these resonances and the nature of the broadening process are of great importance. The situation in highly polar GaN is more challenging than any other semiconductor due to much stronger coupling between LO phonons and excitons. In this paper, we report the dependence of the photoluminescence (PL) linewidth on temperature in GaN epilayers grown by low-pressure metal-organic chemical-vapor deposition (LP-MOCVD). The linewidth broadening mechanism in this system is under debate. Previously, excitonic linewidths in GaN have been studied by time-integrated and spectrally resolved degenerate four-wave mixing (TI and SR DFWM),[2] absorption,[3] PL,[4] reflectance,[5] electroreflectance,[6] spectroscopic ellipsometry,[7] and thermomodulation.[8] Different authors have considered different origins for the linewidth, and the broadening parameters also scan a wide range. In particular, it is important to understand the exciton linewidths in photoluminescence since the *photonic devices* are based on *emission phenomena*. Nakamura and co-workers[4] have analyzed the linewidths by considering the LO phonons and the low-frequency branch of the E_2 mode. They could not obtain a satisfactory fitting of the linewidths by taking into account acoustic-phonon scattering. In contrast, our findings have shown that exciton–acoustic-phonon interactions do contribute to the linewidth and they must be considered. We compared our broadening parameters of free excitons *A* and *B* in GaN with those obtained by different experimental techniques in the literature. We determined the binding energy of free excitons *A* and *B* from the temperature dependence of the photoluminescence intensities. We also discussed the exciton-phonon interactions in comparison with wide-band-gap II-VI (Refs. 9–14) and III-V (Refs. 15–17) semiconductors and their importance in the realization of exciton-based photonic devices. In fact, the lasing mechanism in GaN-based devices—excitonic versus band-to-band recombination—has not been understood so far and this topic is currently under intense debate.[1] Therefore, the properties of excitons in these systems, particularly those which depend on temperature, are very important since they can shed some light on some of the critical issues.

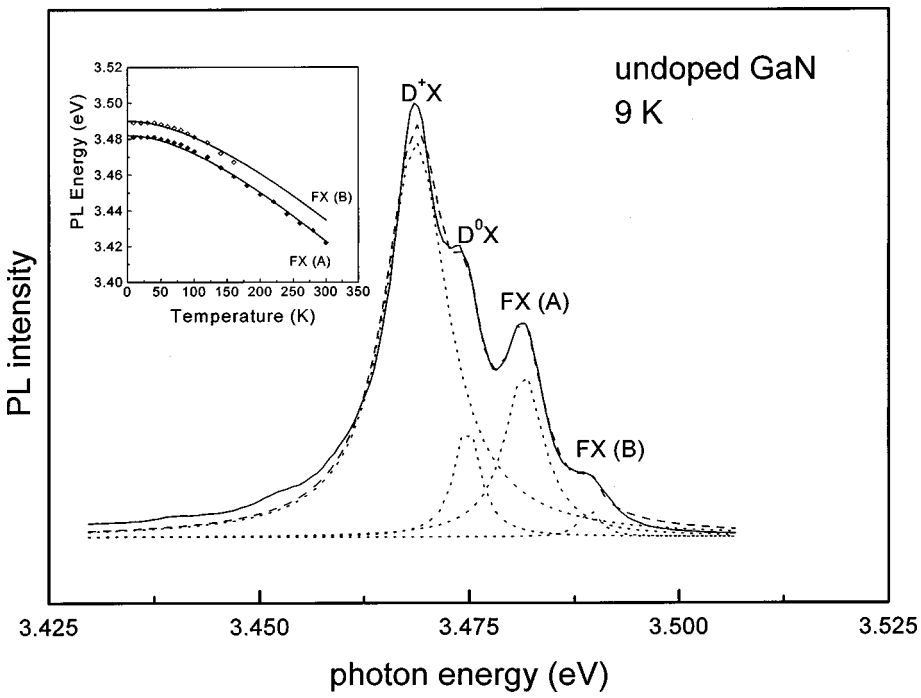

FIG. 1. Luminescence spectrum of GaN epilayer at 9 K. Dotted lines show the deconvolution into Lorentzian fitting of various transitions. Dashed line is the Lorentzian fitting of the experimental spectrum. The inset shows the temperature dependence of the transition energies of free excitons *A* and *B*.

The experimental details were given in our previous reports.[18,19] Figure 1 shows the 9 K luminescence spectrum of a GaN epilayer on a *c*-face sapphire substrate. The spectrum has four unresolved and overlapping peaks. We have done the Lorentzian fitting in order to obtain the peak positions and linewidths accurately. The dotted lines show the deconvoluted bands and the dashed line shows the Lorentzian fitting of the experimental spectrum. FX (*A*) and FX (*B*) are the free-exciton *A* and *B* transitions with respective energies at 3.481 and 3.489 eV with a peak separation of 8 meV, D^0X is the neutral donor bound exciton with a peak at 3.475 eV, and D^+X at 3.469 eV is interpreted as being due to an ionized donor bound exciton transition. A detailed report on the optical properties of residual shallow donors will be published elsewhere,[18] in which the basis of assignment for the rarely observed ionized donor bound exciton is discussed. Free exciton *A* could be seen up to room temperature, while free exciton *B* was observed up to 150 K. The inset of Fig. 1 shows the temperature dependence of free-exciton transitions which follows the same variation as that of the band gap. This result is another confirmation for the assignment of the transitions to free excitons.

Figures 2(a) and 2(b) show the temperature dependence of the linewidths of the free excitons *A* and *B*, respectively. Solid circles are the experimental linewidths. We have done the fitting of the observed linewidths according to a theoretical model. The line broadening in semiconductors has been considered theoretically by various workers. The basic formalism for phonon-assisted optical absorption and exciton lifetimes was developed by Toyozawa[20] and by Segall and Mahan.[21] It was applied to describe absorption and emission in a number of materials by Segall.[22] In this formalism, light absorption by an exciton was discussed in the context of linear-response theory. The absorption is given by an imaginary part of the exciton Green's function and has a Lorentzian shape. The temperature dependence of the width of the lowest $1S$ exciton in semiconductors was initially reported by Segall[23] and the most comprehensive treatment was later given by Rudin, Reinecke, and Segall.[24] According to them, the exciton linewidth $\Gamma(T)$ at any temperature T can be written as

$$\Gamma(T)=\Gamma_0+\gamma_{\mathrm{ph}}T+\Gamma_{\mathrm{LO}}N_{\mathrm{LO}}(T). \tag{1}$$

In high-purity crystals the linewidth originates from interactions with thermal phonons, and in polar semiconductors it is dominated by LO-phonon broadening. But in practice the semiconductor grown by any method has some background impurities, crystal imperfections, and they contribute to inhomogeneous broadening. This is represented as Γ_0 in the above equation. At very low temperatures, phonons are not active and the contribution to the linewidth is mainly from inhomogeneous broadening. The best way to obtain Γ_0 is by extrapolation of the linewidth to 0 K. The second and third terms on the right-hand side of Eq. (1) are the homogeneous linewidths and are due to acoustic-phonon and LO-phonon scatterings, respectively. The processes involving acoustic phonons are only intraband scattering of excitons. The contribution due to acoustic phonons increases linearly with temperature T and is represented as $\gamma_{\mathrm{ph}}T$, where γ_{ph} is the exciton–acoustic-phonon coupling strength. The last term in Eq. (1) arises from interactions with LO phonons and is proportional to the Bose function $N_{\mathrm{LO}}(T)$ for LO-phonon occupation, which is equal to $1/[\exp(h\nu_{\mathrm{LO}}/kT-1)]$, where $h\nu_{\mathrm{LO}}$ is the LO-phonon energy in GaN and Γ_{LO} represents the strength of exciton–LO-phonon coupling. Interactions of excitons with LO phonons are described by the Fröhlich inter-

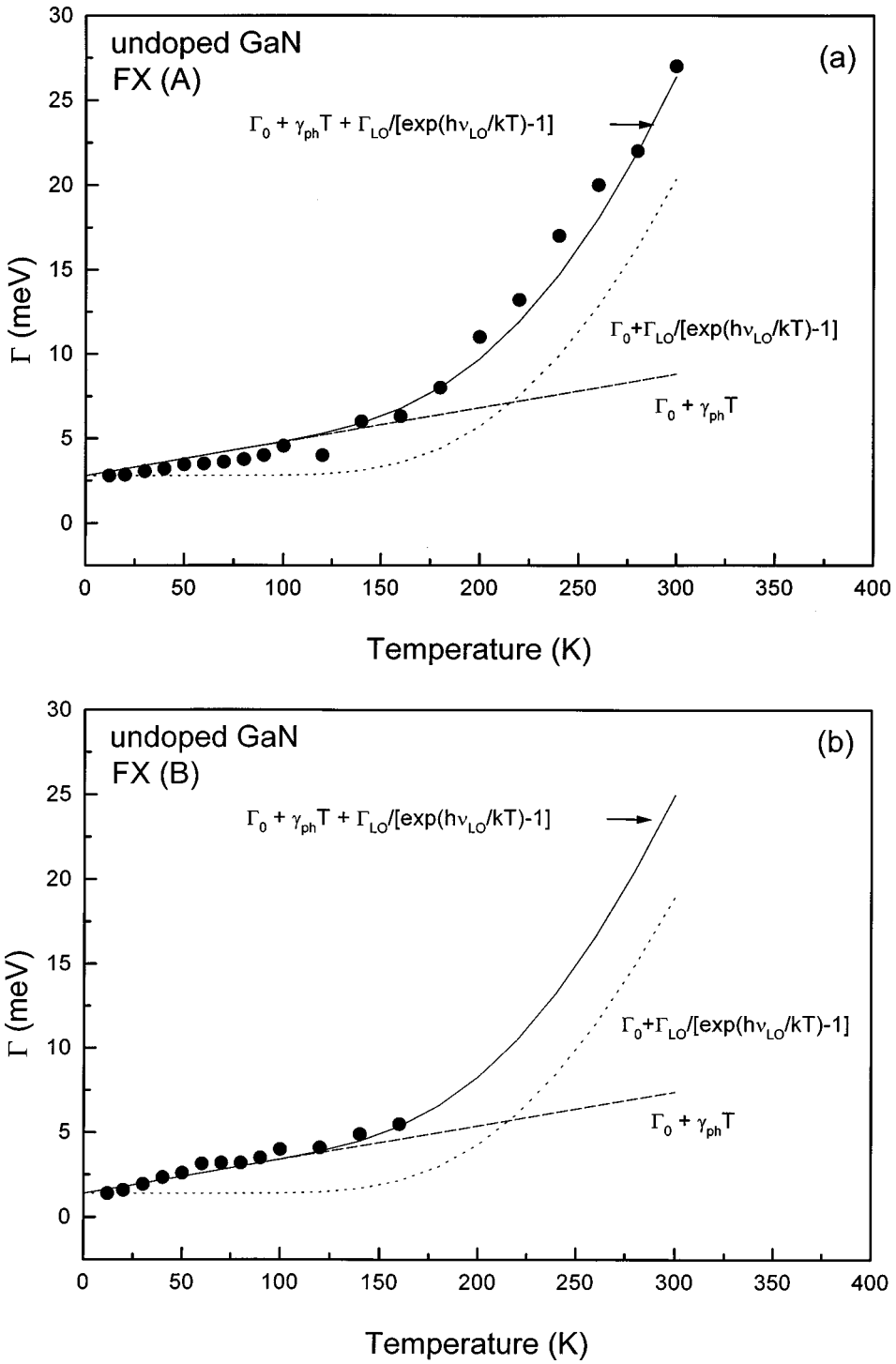

FIG. 2. Temperature dependence of the linewidths of (a) free exciton *A* and (b) free exciton *B* in GaN. Solid circles are the experimental points. Solid curve is the fitting of the experimental linewidths considering acoustic-phonon scattering and LO-phonon scattering. Dashed lines show the contribution from the acoustic-phonon scattering alone, and dotted lines show the contribution from the LO phonons only. In both the cases inhomogeneous broadening was added.

action. Phonons can scatter the exciton to the same bound state or higher-lying bound states.

In Fig. 2 the solid lines are the least-squares fitting considering acoustic-phonon scattering and LO-phonon scattering, in addition to inhomogeneous broadening. We have taken the LO-phonon energy of 91.5 meV.[25,26] The hexagonal GaN crystallizes in the wurtzite structure belonging to the $C_{6v}(P6_3mc)$ space group. There are two formula units per primitive cell and all the atoms occupy the sites of symmetry C_{3v}. In Raman spectroscopy six optical modes $1A_1$ (TO), $1A_1$ (LO), $1E_1$ (TO), $1E_1$ (LO), and $2E_2$ at 533, 735, 561, 743, 144, and 569 cm^{-1} were observed. We find the fit to be excellent with the following parameters: $\Gamma_0 = 2.8$ meV, $\gamma_{ph}=21$ μeV/K, and $\Gamma_{LO}=525$ meV for FX(A) and $\Gamma_0=1.4$ meV, $\gamma_{ph}=22$ μeV/K, and $\Gamma_{LO}=495$ meV for FX(B). In Fig. 2 the dashed lines show the contribution from

TABLE I. Linewidth broadening parameters for free-exciton transitions in GaN.

Technique	Γ_{LO} (meV)	γ_{ph} (μeV/K)	Reference
Photoluminescence of free exciton *A*	525	21	This work
free exciton *B*	495	22	This work
Spectrally resolved degenerate four-wave mixing	470	13	2
Time-integrated degenerate four-wave mixing	390	16	2
Absorption of free exciton *A*	375	15	3
Photoluminescence of free exciton *A*	165		4
Reflectance of free exciton *A*	208	15.3	5
Electroreflectance of free exciton *A*	60		6
Electroreflectance of free exciton *B*	74		6
Spectroscopic ellipsometry	104		7
Thermomodulation spectroscopy	179		8

acoustic-phonon scattering and the dotted lines show the contribution from the LO phonons to the linewidth. In all cases, the inhomogeneous broadening is also added. It is easily noticeable that the acoustic phonons contribute up to 120 K very significantly, but the contribution from LO phonons is negligible up to this temperature. From 120 K onwards the participation of LO phonons causes the linewidth to increase sharply. At 200 K there is a crossover and the LO phonons dominate over the acoustic phonons.

The value of the exciton–LO-phonon coupling parameter, Γ_{LO}, deduced by us is very high. It indicates very strong interaction between excitons and LO phonons. The main reason for a high value of Γ_{LO} is that GaN is a polar material with a very high Fröhlich constant.[27] The exciton-phonon interactions manifest in two ways in the optical spectra. First is the line broadening of the absorption or emission spectra as discussed thus far. Second is the appearance of phonon-assisted transitions in the absorption and emission spectra. The phonon replicas of free-exciton transitions in photoluminescence have been observed by some of us[28] in the epitaxial layers of GaN grown by rotating disk MOCVD. Shan *et al.*[29] observed very clear features of phonon replicas in the photoluminescence excitation (PLE) spectroscopy with LO-phonon energy of 91.5 meV and also a small bump in the absorption spectra with a similar energy spacing, which they attributed to a LO-phonon replica, while Kovalev *et al.*[30] reported phonon replicas in the PLE and PL of GaN epilayers grown on sapphire as well as SiC. In Mg-doped GaN grown by rotating disk MOCVD, we also observed up to four phonon replicas of the donor-acceptor pair transitions.[31] All these experimental results strongly support the very high value of Γ_{LO} obtained by us, indicating a very strong coupling between excitons and LO phonons of the GaN lattice.

Table I compares the broadening parameters for GaN obtained by different experimental techniques. The only other result on the photoluminescence linewidths was reported by Nakamura and co-workers.[4] But they could not get a satisfactory fitting of the linewidths by considering the LO-phonon and acoustic-phonon scattering. So they have considered the low-frequency (144 cm^{-1}) branch of the E_2 mode along with LO-phonon coupling to fit their data. The clear-cut difference between our interpretation and Nakamura's conclusion was that our analysis has shown the participation of acoustic phonons in the low-temperature regime while Nakamura *et al.* proposed the E_2 mode interaction for the low-temperature linewidth. As can be seen in Table I, the reported values of Γ_{LO} span a wide range. Our fitted values agree very well with DFWM results while some of the other published results have given very low values. We give below the arguments for this discrepancy.

Pollack and co-workers have reported[6] Γ_{LO} values of 60 and 74 meV. We believe that these are totally incorrect. In their reflectance spectra *A* and *B* excitons were not resolved even at their lowest temperature of measurement. This might have caused unavoidable errors in the estimation of the linewidth. They have shown that the linewidth was constant up to 150 K (please see Fig. 3 in Ref. 6), which is absurd considering the thermally active phonons in GaN. This result shows that their values of linewidths are completely incorrect. They obtained a value of 731 K (63 meV) for the LO-phonon temperature, which does not match with any known LO-phonon energies, which are around 92 meV. The inhomogeneous broadening of 15 meV obtained by them is very high and is perhaps due to poor sample quality. They have used the Bose-Einstein-type expression, which considers only LO-phonon scattering, and acoustic-phonon contributions to the linewidth are not taken into account.

The Γ_{LO} value (104 meV) reported by Petalas *et al.*[7] from the spectroscopic ellipsometry is also low and our impression is that it is incorrect. They made the linewidth measurements from 120 K onwards only and hence they did not have the low-temperature data to make the fitting and analyze the contributions in that temperature region. They also observed the linewidth to be almost constant up to a high temperature of 200 K, which is a wrong result. This shows that the magnitudes of their linewidths were incorrect. The inhomogeneous broadening value of 35 meV of Γ_0 reported by this group was quite high, which shows that their sample quality may not be good or the method of measurement is not right. They made use of a Bose-Einstein-type expression, which considers only LO-phonon scattering, and acoustic-phonon contributions to the linewidth are completely ignored.

In addition to these workers, Li *et al.*,[8] who analyzed the linewidths from modulation spectroscopy, have also used the Bose-Einstein-type expression which considers LO-phonon scattering alone and acoustic-phonon contributions to the linewidth are taken as zero. We believe that in all these cases the linewidths are determined incorrectly (as evidenced by

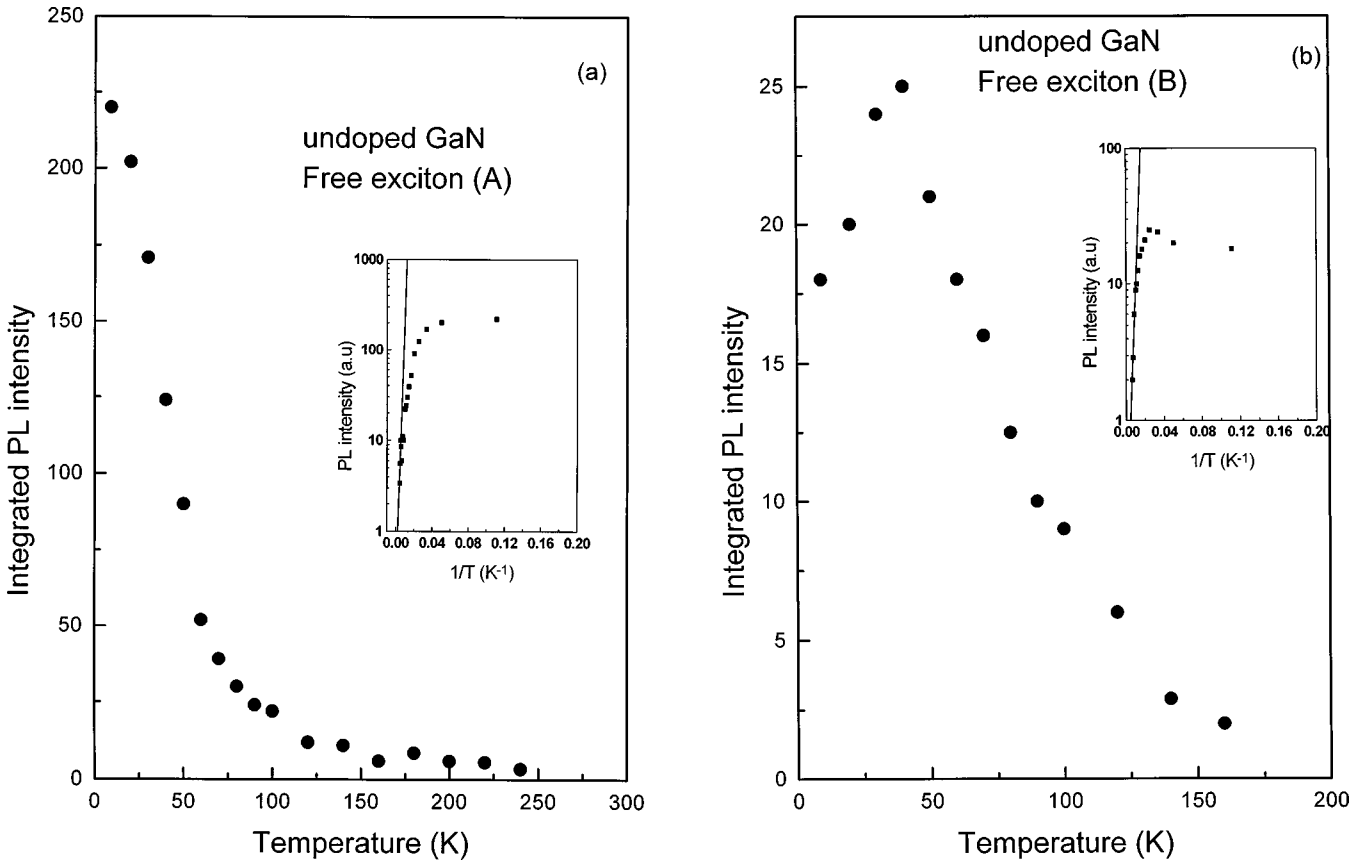

FIG. 3. Temperature dependence of integrated PL intensity of (a) FX (*A*) and (b) FX (*B*). The insets show the activation energy plots.

the fact that they are constant up to 150–200 K), which has led to low values of the fitting parameters.

It is remarkable that our values of broadening parameters agree well with those reported by Fischer *et al.*[2] on the SR and TI DFWM experiments on GaN, who obtained a value of 470 meV for Γ_{LO} and 13 μeV/K for γ_{ph}. Perhaps the most accurate determination of the exciton-phonon coupling parameter is from the spectrally resolved degenerate four-wave mixing. In these measurements the excitonic contribution is several orders of magnitude larger than the free-carrier contribution. The results obtained from other experiments such as absorption contain errors due to significant contributions to the linewidth from free carriers. We believe that the linewidths reported from all the other techniques are not accurate since they do not detect the exciton transition directly. For example, in spectroscopic ellipsometry one measures the complex dielectric function ε which is related to the point density of states for interband transitions, while in the reflectance experiment the measurement of ε itself is an indirect method. Considering our own measurements and those of Fischer *et al.*, we believe that the lower limit of Γ_{LO} must be around 450 meV. Such a high value is mainly attributed to the very intense Fröhlich interaction in GaN. The evidence and ramifications of Fröhlich interactions were already discussed in previous sections

We compare the broadening parameters in GaN with those in wide-band-gap II-VI and other III-V semiconductors. For ZnSe, Γ_{LO} value was reported to be 81 meV from the degenerate four-wave mixing experiments.[9] In the case of a CdTe/CdZnTe single quantum well, a high value of 400 meV for Γ_{LO} was reported[11] from the photoluminescence experiments. However, the values given by others for CdZnTe-type single and multiple quantum wells were in the range of 16–25 meV from absorption and luminescence studies.[12–14] For GaAs and $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ bulk and quantum wells, Γ_{LO} values obtained from absorption/reflectance were very small in the range of 5–10 meV.[15–17] The trend in the Γ_{LO} value can be understood by considering the polarity of the material which increases in the order of GaAs, ZnTe, ZnSe, and GaN. The energy of LO phonons also has a profound effect on the Γ_{LO} value. It may be noted that the $h\nu_{\mathrm{LO}}$ for GaN is very high (92 meV) compared to other semiconductors such as GaAs (36.8 meV), ZnTe (25.5 meV), ZnSe (30.5 meV), CdTe (21.2 meV), and CdSe (26.1 meV).[24]

Now we discuss the acoustic-phonon coupling parameter γ_{ph}. Most of the other workers[4,6–8] did not even consider the contributions of acoustic phonons in GaN. The importance of acoustic phonons in linewidth broadening has also been theoretically treated by Lee, Koteles, and Vassell,[32] who concluded that acoustic phonons dominate up to 120 K, which agrees with our findings. Our γ_{ph} value agrees reasonably well with the values reported by others[2,3,5] for GaN. Exciton–acoustic-phonon interactions occur via the deformation potential and piezoelectric interaction. The observed small differences in the γ_{ph} value can be explained as being due to differences in the deformation potential in different samples. The importance of exciton–acoustic-phonon scattering has also been addressed by Shah and co-workers[33] recently in their time-resolved DFWM experiments on GaAs quantum wells. It is interesting to compare the γ_{ph} value in GaN with other III-V and II-VI semiconductors. The γ_{ph}

values were reported as 4.6 μeV/K for GaAs quantum wells, 11.0 μeV/K for ZnSe bulk as well as multiple quantum wells (MQW) of ZnCdSe/ZnSe,[9] 13.7 μeV/K for MQW of CdTe/CdMnTe.[10] This comparison shows that the γ_{ph} value is in the same range for all the semiconductor materials. This is because γ_{ph} depends only on the deformation potentials and the Fröhlich constant has no effect on it. These comparisons also prove that only Γ_{LO} is very much influenced by the intensive Fröhlich interactions in GaN. The treatment given by us to understand the exciton linewidths has also been applied very recently to investigate the influence of quantum confinement on the magnitudes of acoustic- and optical-phonon scattering in ZnCdSe/ZnSe MQW (Ref. 34) and $In_xGa_{1-x}As/GaAs$ single quantum wells[35] by TI-DFWM experiments. This gives additional support to the approach used by us.

Figure 3(a) shows the temperature dependence of integrated PL intensity of free exciton *A* and the inset shows the same quantity as a function of $1/T$. From the slope of the straight line, the activation energy of the free exciton is estimated as 26 meV. This can be taken as *A* exciton binding energy. For free exciton *B* the PL intensity initially increases upto 40 K and then decreases due to nonradiative relaxation, as shown in Fig. 3(b). The initial increase in intensity occurs since the *B* exciton is at a higher energy than the *A* exciton. The binding energy of FX (*B*) is also evaluated as 26 meV. This value of binding energy agrees reasonably well with the values estimated recently[31,32] from the ground and excited spectra in the photoreflectance.

In order to achieve room-temperature photonic devices, it is necessary to have excitons which can exist up to room temperature. The existence of room-temperature excitons depends upon the delicate balance between two competing factors. The first is the exciton–LO-phonon interaction parameter and the second is the exciton binding energy. A very large value of Γ_{LO} will make the exciton line broadened, so much so that at high temperatures the line will vanish completely. But the larger binding energy of the exciton can resist the phonon interaction and it is possible to observe room-temperature excitons. Indeed this is what happens in the case of GaN, which has a large binding energy of 26 meV. In order to realize stable room-temperature devices, we would like to suggest here that it is better to work with quantum confined systems of GaN, which have higher binding energy and lower LO-phonon coupling as a direct consequence of quantum phenomena. In addition, quantum wells also have much smaller inhomogeneous broadening.

In conclusion, we have reported the temperature dependence of the linewidth of free exciton *A* and *B* and proposed the emission broadening mechanism. It was shown that acoustic-phonon scattering must also be considered in addition to LO-phonon scattering. The broadening parameters were compared with those obtained by different experimental methods and also with other III-V and II-VI wide-band-gap semiconductors. The importance of linewidth and binding energy of excitons in achieving room-temperature photonic devices was discussed.

One of the authors (A.K.V.) thanks the Brain Pool program of Korea for support. This research was financially supported by the Creative Research Initiative of the Ministry of Science and Technology of Korea.

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