Spectral features of LO phonon sidebands in luminescence of free excitons in GaN

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In the paper a combined experimental and theoretical investigation of the longitudinal optical phonon sidebands (PSBs) in the luminescence of free excitons in GaN at moderately high temperatures was reported. The spectral features, including line broadening, shift, and asymmetry of the one- and two-phonon PSBs, were revealed both experimentally and theoretically. It is found that the linewidth of the one-phonon PSB is surprisingly always larger than that of the two-phonon PSB in the interested temperature range. Moreover, the thermal broadening rates of the one- and two-phonon PSBs are considerably different. We adopted the Segall–Mahan theory [B. Segall and G. D. Mahan, Phys. Rev. **171**, 935 (1968)] to compute the PSB spectra of the free excitons in GaN. Only one adjustable parameter, the effective mass of the holes, was used in the calculations. For the one-phonon PSB, an excellent agreement between theory and experiment is achieved when an adequate effective mass of the holes was used. © *2005 American Institute of Physics*.
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I. INTRODUCTION

GaN is a wide direct-gap III-V semiconductor and usually crystallizes in a wurtzite structure. Although these basic properties of GaN were known much earlier, extensive investigations about GaN have been undertaken only in the past decade since the first successful demonstration of its application in short-wavelength visible light-emitting devices.[1] The current status in the study of GaN is that the empirical development of GaN-based materials and devices is advancing rapidly and that fundamental understanding is struggling to catch up.[2] This is particularly true in the aspect of exciton–phonon or electron–phonon interaction in spite of its fundamental importance to the optical and electrical properties of GaN and even to the performance of GaN-based devices. Due to the ionic nature of GaN, the Fröhlich-type longitudinal optical (LO) phonon–exciton interaction is the strongest. As a result of such interaction, the LO phonon sidebands (PSBs) appear in the luminescence spectra of excitons. The PSBs of free and bound excitons were observed by Dingle *et al.* in 1971.[3] However, a systematic study of the PSBs and the relevant exciton–phonon interaction issues in GaN was not conducted until recent years. The available experimental investigations[4–12] of the PSBs of excitons in GaN have significantly enriched the knowledge of the exciton–phonon interactions. In these studies, most experimentalists employed a simple empirical expression for the transition probabilities of the LO phonon-assisted radiative recombination of free excitons, suggested by Permogorov,[13] to fit the observed

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spectral line shape.[4–6,8,9,11,12] A further understanding of the PSBs and related problems clearly requires a detailed and rigorous calculation, which is still lacking. By introducing an extended Green's-function approach, Segall and Mahan[14] developed a more rigorous theory for computing the phonon-assisted radiative recombination of free excitons in compound semiconductors. They also showed that the Permogorov's empirical expression is essentially correct around the threshold. In this work, we employ the Segall–Mahan theory to calculate the LO phonon-assisted excitonic emission spectra in GaN at several typical temperatures, and then directly compare the theoretical curves with the experimental data.

In Sec. II we briefly describe the experiment, while the theoretical calculation is given in Sec. III. In Sec. IV we compare and discuss the experimental data and calculated curves. In the final section, the conclusion is given.

II. EXPERIMENT

The experimental sample was a 2.88-μm-thick GaN epilayer grown on sapphire (0001) with metalorganic vapor phase epitaxy. In the photoluminescence measurements, the sample was mounted on the cold finger of a Janis closed-cycle cryostat with varying temperatures from 3.5 to 300 K and excited by the 325-nm line of a Kimmon He–Cd continuous-wave laser with output power of 37 mW. The emission signal from the sample was dispersed by a Spex 750M monochromator and detected with a Hamamatsu R928 photomultiplier. The data acquisition, record, and monochromator control were performed with a PC. Notice that no lock-in amplifier was used because of a strong enough emission signal, which also indicates the high quality of the sample.

III. THEORETICAL CALCULATION

As mentioned earlier, we adopted the Segall–Mahan rigorous theory to compute the PSBs of free excitons in GaN. Based on the Green's-function approach, the Segall–Mahan theory considered the effects of exciton–phonon interactions at all orders and thus included the line broadening, shift, asymmetry, and all renormalizations. Now we rewrite the major equations of the Segall–Mahan theory for phonon-assisted radiative recombination of free excitons.

The Hamiltonian for the coupled system involving excitons, phonons, and photons can be written as

$$H = H_0 + H_{\text{ex-R}} + H_{\text{ex-L}}, \tag{1}$$

where H_0 is the unperturbed part, $H_{\text{ex-}L}$ and $H_{\text{ex-}R}$ are coupled terms between excitons and LO phonons and between excitons and photons, respectively. Following the Segall–Mahan notation, the terms on the right side of Eq. (1) are expressed by

$$H_0 = \sum_{\lambda K} \varepsilon_{\lambda K} c^+_{\lambda K} c_{\lambda K} + \sum_q \omega_q a^+_q a_q + \sum_K \omega_K \alpha^+_K \alpha_K, \tag{2a}$$

$$H_{\text{ex-}R} = \sum_{\lambda K} \frac{M_\lambda(K)}{\sqrt{\omega_K}} [c^+_{\lambda K} \alpha_K + \alpha^+_K c_{\lambda K}], \tag{2b}$$

and

$$H_{\text{ex-}L} = \sum_{\lambda,\lambda';K,q} V(q)_{\lambda\lambda'} c^+_{\lambda,K+q} c_{\lambda' K} (a_q + a^+_q). \tag{2c}$$

Here, $c^+_{\lambda K}(c_{\lambda K})$, $a^+_q(a_q)$, and $\alpha^+_K(\alpha_K)$ are the creation (annihilation) operator for excitons with momentum K and quantum number λ, phonons of wave vector q, and photons of wave vector K, respectively. $\varepsilon_{\lambda K}$, ω_q, and ω_K are the corresponding energies, $M_\lambda(K)$ is the exciton–photon matrix element, and $V(q)$ is the coupling strength between excitons and LO phonons.

In the Segall–Mahan theory, the absorption coefficient is calculated first and then the emission probability is found using the following relationship:

$$W_{\text{em}}(h\nu) \propto e^{-h\nu/k_B T} W_{\text{abs}}(h\nu). \tag{3}$$

For the exciton absorption assisted by one LO phonon, the absorption coefficient has been derived as[15]

$$\alpha_{1\text{ph}}(h\nu) = \frac{4\pi\beta_{A,1}}{4a(\varepsilon')^{1/2}} \frac{e^2}{\hbar c} \frac{\hbar\omega_l}{B} (\varepsilon_\infty^{-1} - \varepsilon_s^{-1}) \left(\frac{E_{A,1}}{E_{A,1} - h\nu}\right)^2 \times \left[\frac{M_\parallel B}{\mu_\perp \Delta}\right]^{1/2} N(\hbar\omega_l) I(h\nu), \tag{4}$$

with $N(\hbar\omega_l) = (e^{\hbar\omega_l/k_B T} - 1)^{-1}$.

The integral $I(h\nu)$ is given by

$$I(h\nu) = \int_0^1 dx \left[1 + \frac{M_\parallel - M_\perp}{M_\perp} x^2\right]^{-1} \times \left\{ \frac{2}{S_e} \frac{(1+\eta)^2 - S_e^2}{[(1+\eta)^2 + S_e^2]^2} \text{Im}\, F_e(\eta) + \frac{4(1+\eta)}{[(1+\eta)^2 + S_e^2]^2} \text{Re}\, F_e(\eta) - [e \to h] \right\}^2, \tag{5}$$

with Re and Im denoting the real and imaginary parts and

$$F_e(\eta) = {}_2F_1(2, 1 - \eta^{-1}, 2 - \eta^{-1}, [1 - \eta^2 + S_e^2 - i2S_e\eta]/[(1+\eta)^2 + S_e^2]).$$

Here, ${}_2F_1$ is the hypergeometric function and $[e \to h]$ denotes the term obtained from the previous bracketed terms by interchanging the e and h subscripts. The definitions of S_e, S_h, η, a, $\beta_{A,1}$, B, $M_\parallel$, $M_\perp$, $\mu_\perp$, and $E_{A,1}$ can be found in Ref. 15.

For transitions of free excitons assisted by two LO phonons, the absorption coefficient is given by[16]

$$\alpha_{2\text{ph}} = \frac{\varepsilon_s}{\pi a} \frac{4\pi\beta}{(\varepsilon')^{1/2}} \left(\frac{e^2}{\hbar c}\right) \left(\frac{M}{4\mu}\right)^3 \left(\frac{\hbar\omega_l}{B}\right)^2 (\varepsilon_\infty^{-1} - \varepsilon_s^{-1}) \times \left(\frac{E_{x1}}{E_{x1} - h\nu}\right)^2 \left(\frac{\langle E_{xn'}\rangle - E_{x1}}{\langle E_{xn'}\rangle - h\nu}\right)^2 N^2(h\omega_l) \sum_n I_n(h\nu), \tag{6}$$

where

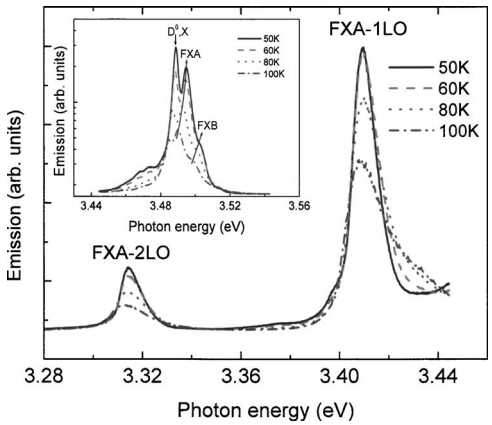

FIG. 1. Measured luminescence spectra of free excitons in GaN at several representative temperatures. The inset figure represents the resonant emission spectra of various excitons.

$$I_n(h\nu) = \int_0^\infty \frac{dz}{z} \int_{|\beta_n - z|}^{\beta_n + z} \frac{dz'}{z'} |[1|U(2za^{-1})|1][1|U(2z'a^{-1})|n] \times [(b+z^2)^{-1} - (d+z^2)^{-1}] + (d+z^2)^{-1} \times [1|U(2za^{-1})U(2z'a^{-1})|n] + (z \to z')|^2,$$

with

$$b = (E_{x1} - \hbar\omega_l - h\nu)M/4\mu B,$$

$$d = (\langle E_{xn''}\rangle - \hbar\omega_l - h\nu)M/4\mu B,$$

$$\beta_n = \frac{1}{2} x_n a,$$

and

$$[n, l=0|U|n=1] = \left\{ S_1(p_h) + n^{-2}\left[\frac{1}{4} S_3(p_h) - \frac{1}{12} S_4(p_h) \right] \right\} - \{p_h \to p_e\},$$

where $p_j = qam_j/M$ $(j = e \text{ or } h)$,

$$S_k(p) = -p^{-1} 2^{k+1} (1+p^2)^{-k-1} \exp[-2/(1+p^2)] \times \{\sin[2p/(1+p^2)]\mathrm{Re}(1-ip)^{k+1} + \cos[2p/(1+p^2)]\mathrm{Im}(1-ip)^{k+1}\},$$

with Re and Im denoting the real and imaginary parts.

IV. COMPARISON BETWEEN EXPERIMENT AND THEORY AND DISCUSSION

Figure 1 shows the measured photoluminescence (PL) spectra on the low-energy side of the zero-phonon lines of the GaN sample at several typical moderately high temperatures. The inset figure represents corresponding zero-phonon lines. At lower temperatures, the zero-phonon line (denoted by D^0,X) of excitons bound at shallow donors dominates.[4,8–10] Correspondingly, the PSBs of the bound excitons are also relatively strong. As the temperature is raised, the bound excitons efficiently dissociate from the donors and become free excitons because of the weak localization and hence the small thermal activation energy of the bound excitons. At about 80 K and above, the zero-phonon line [denoted by free exciton *A* transition (FXA)] of the free excitons is stronger than that of the bound excitons. However, earlier at 50 K, the PSBs of the bound excitons becomes almost unobservable due to the relatively small coupling strength of the bound excitons with the LO phonons.[9,12] When the temperature is higher than 100 K, the PSBs of free electrons and holes appear.[11] Therefore, we investigate the PSBs of the free excitons in the temperature range from 50 to 100 K.

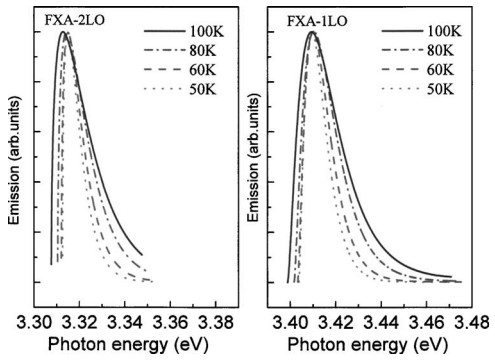

FIG. 2. Calculated one- and two-phonon-assisted luminescence spectra of free excitons. Note that all the curves are normalized.

From Fig. 1, one can see the asymmetric line shape of both the one- and two-phonon PSBs. Moreover, the asymmetry of both lines increases with increasing temperature. These results show that the asymmetric line shape of the PSBs is likely an intrinsic property of the exciton–phonon interaction. Another distinct feature in the spectra of Fig. 1 is the rapid broadening of PSB peaks as the temperature increases. A careful look at the spectra enables us to find that the temperature dependences of the one-phonon and two-phonon PSBs are likely to be different from each other. For example, the broadening rate of the one-phonon PSB with temperature seems higher than that of the two-phonon one. It is thus reasonable for us to speculate that the lattice temperature and the kinetic energy of excitons may have a greater influence on the one-phonon PSB than on the two-phonon one. As mentioned earlier, a theoretical calculation based on a rigorous model is obviously needed in order to understand these observed spectral features of the PSBs.

Figure 2 shows the calculated PSB spectra with the Segall–Mahan theory briefly described in Sec. III. The values of various parameters adopted in the calculation are listed in Table I. The theoretical spectra clearly show the asymmetric line shape of the PSBs, which indicates that the asymmetry of the PSBs is a characteristic of the exciton–phonon interaction. Toyozawa theoretically explained the physical origin causing the asymmetry of the PSBs. As pointed out by Toyozawa, the quantum interference between the intermediate states of excitons via LO phonons leads to the asymmetric line shape of the phonon-assisted optical transition.[19] In fact, Segall and Mahan already proved that their theory is equivalent to a main expression derived by Toyozawa under the appropriate conditions, such as sufficiently separated bands of exciton. For the GaN case studied in the present work, the binding energy of the free exciton is about 25 meV.[11] The

TABLE I. Parameters used in the calculation of the absorption coefficient.

Parameters	Symbol	GaN
Electron effective mass (units of m_0)	m_e	0.2[a]
Hole effective mass (units of m_0)	$m_{h\perp}$	0.5
	$m_{h\parallel}$	0.6
Exciton binding energy (eV)	B	0.0254[b]
LO phonon energy (meV)	$\hbar\omega_l$	91.2[c]
Statistic dielectric constant	$\varepsilon_s=(\varepsilon_\perp\varepsilon_\parallel)^{1/2}$	8.9[c]
High-frequency dielectric constant		5.35[c]
	$\varepsilon_\infty=(n_\perp n_\parallel)^{1/2}$	
Dielectric constant at E_{A1} in the absence of $n=1$ exciton	ε'	8.0
$A(n=1)$ exciton position at T K(eV)	50 K, $E_{A,1}=3.4949$	
	60 K, $E_{A,1}=3.4944$	
	80 K, $E_{A,1}=3.4928$	
	100 K, $E_{A,1}=3.4900$	
Zero-frequency polarizability due to $n=1$ A,B, or C	0.0066	
exciton $(\varepsilon\perp c)4\pi\beta_{A,1}$	0.0227	
	0.030	

[a]Reference 18.
[b]Reference 11.
[c]Reference 17.

excited states of free excitons in GaN can be clearly resolved in the luminescence spectrum at low temperature. The well-separated exciton bands provide the intermediate states for the optical transitions involving phonons. For the luminescent processes, these transitions via the intermediate states consist of a continuous background which interferes with the discrete transition at $E_{1S}-\hbar\omega_{\mathrm{LO}}$ (E_{1S} is the lowest-lying state of exciton and $\hbar\omega_{\mathrm{LO}}$ is the LO phonon energy). Such interference produces a typical asymmetric line shape first derived by Fano when he theoretically dealt with the scattering of electrons by helium nuclei.[20] Therefore, the observed asymmetric line shape of the PSBs is from the phonon-induced Fano-type quantum interference.

Another spectral feature in the theoretical spectra shown in Fig. 2 is the rapid broadening of the lines with temperature. It is known that the increase of the lattice temperature will lead to an increase of the average center-of-mass kinetic energy of excitons which changes the distribution of exciton statistics.[21] The influence of the exciton's thermal distribution on the PSBs is mainly reflected by the Boltzmann factor in Eq. (3) relating the absorption and emission. Our theoretical calculations show that the phonon-assisted excitonic absorption is insensitive to the variation of the temperature. It is thus concluded that the observed strong broadening of the PSBs with the temperature is mainly due to the thermal distribution of excitons. In addition, the variation of the kinetic energy of excitons also affects the exciton–phonon scattering character, especially in the first-order scattering process. According to Segall and Mahan's theory, excitons of low kinetic energy or photons of energy slightly above the one phonon threshold are of most interest for luminescence. They showed that for the excitons with energies slightly above the threshold, the first-order phonon-assisted emission probability $P_1(h\nu)$ is

$$P_1 \sim \Delta_1^{3/2} e^{-\Delta_1/k_B T}. \tag{7}$$

The $\Delta^{3/2}$ threshold behavior in the above equation arises from the linear dependence of the scattering matrix element on Δ_1 at small Δ_1 and the $\Delta^{1/2}$ density-of-state factor. Notice that expression (7) is the dependence of the one-phonon emission spectra on the kinetic energy of excitons suggested by Permogorov.[13] Permogorov suggested such a dependence on the basis of the approximate $(3/2)k_B$ value of the slope of the measured one-phonon PSB widths versus the temperature. Indeed, we observed such an approximate value for the one-phonon PSB of free excitons in GaN.[9] For the two-phonon emission, around the threshold, the emission probability $P_2(h\nu)$ is

$$P_2 \sim \Delta_2^{1/2} e^{-\Delta_2/k_B T}. \tag{8}$$

From expressions (3) and (8), we can know that around the threshold the two-phonon-assisted absorption probability is approximately proportional to the density of states.

Figure 3 shows the full width at half maximum (FWHM) height of measured (solid triangles and circles) and calculated PSB lines at different temperatures. The agreement between theory and experiment is very satisfactory considering the uncertainties of some parameter values of GaN. From Fig. 3, it is seen that the one-phonon line tends to be slightly broader than the two-phonon peak. This is consistent with

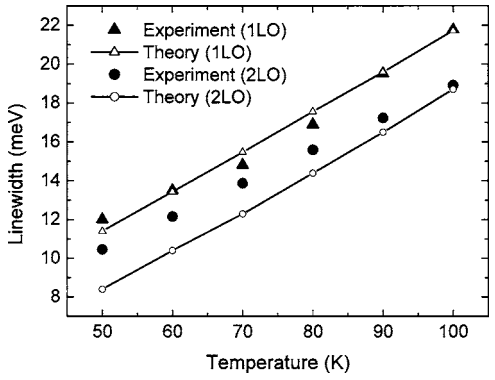

FIG. 3. Measured (solid triangles and circles) and calculated linewidths of the one- and two-phonon PSBs against temperature.

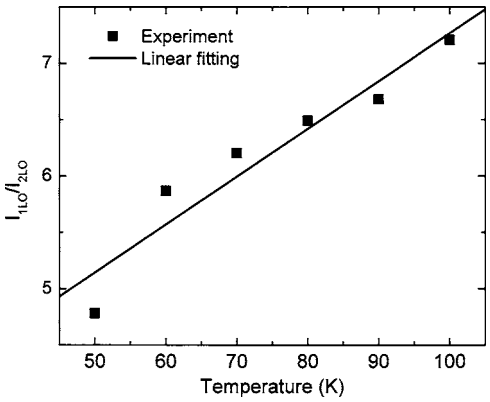

FIG. 4. Measured integrated intensity ratio of one- and two-phonon lines vs temperature. The solid line is the best linear fitting curve.

the case of CdS.[14] The reason causing this phenomenon is that $P_1(h\nu)$ is a more rapidly varying function of Δ_1 than $P_2(h\nu)$ is of Δ_2.

From Eqs. (7) and (8), it is easily derived that the ratio of the integrated intensity of the one-phonon PSB lines to that of the two-phonon PSB lines is simply proportional to the temperature.[13,14] The experimental values (solid squares) of I_{1LO}/I_{2LO} were illustrated in Fig. 4. The solid line is the best linear fitting curve. As expected, I_{1LO}/I_{2LO} linearly varies with the temperature.

Finally, we would like to briefly discuss the effect of exciton–photon coupling which was taken into account in the calculations. It is known that the exciton–photon coupling leads to the formation of polaritonic states and special dispersion curves of the polaritons (see Fig. 3 in Ref. 14.). The Segall–Mahan theory accounted for the weak exciton–photon coupling case in which the exciton–photon interaction is treated accurately. For the LO phonon-assisted luminescence process in such case, a polariton is scattered from an initial state above the region of strong coupling (the bottleneck region) to a final state below this region where the polariton is essentially a photolike state. The coupling to the LO phonon is through the exciton component of the polariton. Under these circumstances, there is no appreciable difference between the perturbation theoretical results and the results of this more accurate approach. In other words, the polariton effect has only a weak contribution to the LO phonon PSB lines. In the GaN case studied in the present work, the polariton effect will become substantial only when the lattice temperature is higher than 160 K, according to our calculations. However, the exciton–photon coupling would have its most pronounced effect on the zero-LO phonon line of the free excitons even at low temperatures.

V. CONCLUSIONS

In conclusion, the spectral features of LO phonon-assisted radiative recombination of free excitons in GaN were investigated in detail as a function of temperature. It is shown that the temperature dependences of the one- and two-LO-phonon-assisted optical transitions of the free excitons are quite different. This can be reflected by the different temperature dependences of their spectral parameters such as the linewidth, asymmetry, and peak position. The Segall–Mahan theory, taking both the exciton–phonon interaction and the exciton–photon coupling (polariton) effect into account, has been employed to quantitatively interpret these spectral features. The theoretical results are in excellent agreement with the experimental spectra when an adequate effective mass of the holes was adopted in the calculation, which gives us a deeper and better understanding of the exciton–LO phonon interactions in GaN.

ACKNOWLEDGMENTS

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