Effects of interface roughness on photoluminescence full width at half maximum in GaN/AlGaN quantum wells*

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Low temperature photoluminescence (PL) measurements have been performed for a set of GaN/$Al_xGa_{1-x}N$ quantum wells (QWs). The experimental results show that the optical full width at half maximum (FWHM) increases relatively rapidly with increasing Al composition in the $Al_xGa_{1-x}N$ barrier, and increases only slightly with increasing GaN well width. A model considering the interface roughness is used to interpret the experimental results. In the model, the FWHM's broadening caused by the interface roughness is calculated based on the triangle potential well approximation. We find that the calculated results accord with the experimental results well.

Keywords: GaN/AlGaN quantum wells, interface roughness, optical full width at half maximum

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1. Introduction

The group-III nitrides have attracted much attention in the past few years due to their important applications, such as blue/ultraviolet light-emitting diodes,[1,2] laser diodes,[3] photodetectors,[4,5] and high temperature and high power electronic devices.[6] It is well known that the nitrides have very large spontaneous polarization and piezoelectric coupling constants, which would induce large built-in electrostatic fields in these compounds.[7] The built-in field can exert a substantial influence on the concentration and the distribution of the free carriers in the nitride heterostructure grown along the (0001) orientation. Owing to the advances in the epitaxial crystal growth technique, a variety of optoelectronic devices have been fabricated based on the group-III nitrides' quantum well (QW) structures,[8,9] which have attractive peculiarities. However, the ability of the quantum wells depends on the structural perfection in terms of the uniformity of composition and the abruptness of interfaces, which not only affects the scattering of electrons during the process of electron transportation in the relevant devices,[10] but also broadens the spectral width of the emitting spectrum.[11] In terms of the interface effect on the photoluminescence (PL) property, much attention has been paid to GaAs/AlGaAs[11–13] or CdTe QWs;[14] in comparison, there were few reports about nitrides.[15]

In this paper, we investigate the optical full width at half maximum (FWHM) of a set of GaN/$Al_xGa_{1-x}N$ QWs by employing low temperature PL experiments. To illustrate the variation of the FWHM in the QWs, we use a triangle potential well approximation to calculate the effect of the interface roughness on the PL broadening. The model generates a similar output as the experimental results.

2. Experimental details

The GaN/$Al_xGa_{1-x}N$ QWs studied here were grown on *c*-plane sapphire substrates by metal organic chemical vapor deposition (MOCVD) using triethylgallium, trimethylaluminium, and ammonia as the precursors. Five samples labeled as A to E were prepared. Prior to the growth of the GaN/$Al_xGa_{1-x}N$ QWs, a thin AlN buffer layer was grown on the sapphire substrate, which was followed by an n-type $Al_xGa_{1-x}N$ epilayer. Then, the GaN/ $Al_xGa_{1-x}N$ QWs of 30 periods were deposited on the epilayer. Finally, an n-type $Al_xGa_{1-x}N$ cap layer about 100 nm was covered on the top. In samples A and B, both with a well width of about 2 nm, the Al compositions in the $Al_xGa_{1-x}N$ barriers were $x = 0.2$ and 0.3, respectively. In samples C, D, and E, the Al compositions were maintained at about 40%, while the QW's widths were 2 nm, 3 nm, and 4 nm, respectively. The widths of the $Al_xGa_{1-x}N$ barriers in all samples were 5 nm. For the PL measurement at a low temperature, the samples were mounted on the cold finger of a closed-cycle helium refrigerator. The home-made spectroscopy system consisted of a frequency three-fold Ti: sapphire femtosecond laser with a repetition rate of 76 MHz, providing a laser beam of excitation wavelength about 267 nm and average power 3 mW. The focused spot diameter was less than 100 μm, and the spectral resolution was about 0.03 nm at 300 nm.

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3. Results and discussion

The continuous-wave PL spectra of the five samples measured at 7.3 K are shown in Fig. 1. Two emission peaks labeled as PK1 and PK2 are observed in all the PL spectra. The PK1 is assigned to the PL emission from the AlGaN cap layers because its position is in accordance with the Al composition;[16] while the PK2 originates from the localized exciton recombination in the GaN/ $Al_xGa_{1-x}N$ QWs. By analyzing the spectra in Fig. 1, the FWHMs are directly measured to be 33 meV, 41 meV, 69 meV, 70 meV, and 72 meV, respectively, as shown in Fig. 2. It can be seen from the figure that the PL FWHM increases significantly with increasing Al composition in the barrier, while it increases only slightly with increasing well width. In order to investigate the variation of the PL FWHM with both the barrier height and the well width, a theoretical model considering the interface effect is introduced below.

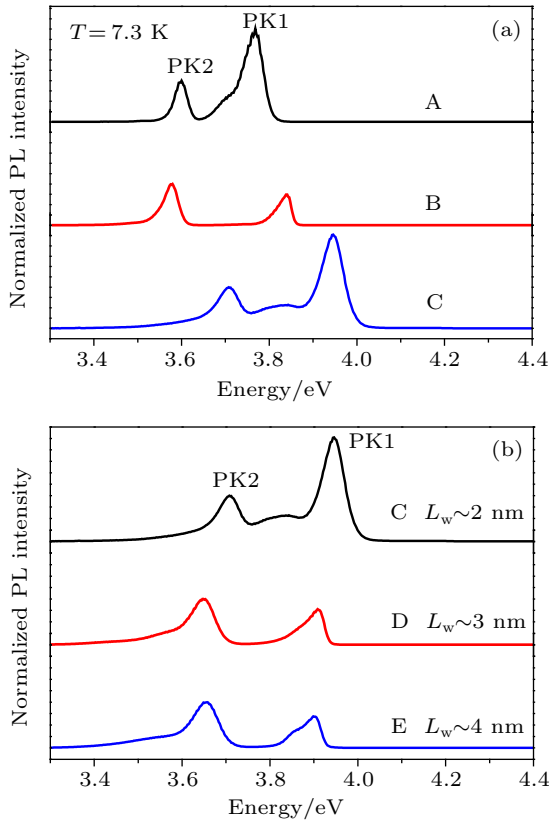


Fig. 1. (color online) PL spectra of GaN/$Al_xGa_{1-x}N$ QW samples at 7.3 K: (a) $x = 0.2$, 0.3, 0.4, with the well width fixed at 2 nm; (b) $x = 0.4$, with the well widths being 2 nm, 3 nm, and 4 nm, respectively. The spectra have been normalized according to PK2 and shifted vertically for clarity.

Generally, the linewidth of excitons in a quantum well at a low temperature can be expressed as

$$\Gamma = \Gamma_{\rm h} + \Gamma_{\rm imh}, \tag{1}$$

where $\Gamma_{\rm h}$ represents the intrinsic linewidth, and $\Gamma_{\rm imh}$ is the extrinsic linewidth. The intrinsic linewidth is mainly determined by several scattering mechanisms, such as phonon scattering[17] and carrier scattering.[18] The $\Gamma_{\rm h}$ should be small under the condition of low temperature and low excitation power. It is well known that $\Gamma_{\rm imh}$ results from the fluctuation of the location potential in space, which is mainly caused by the alloy inhomogeneity or the interface roughness.[12] Since QW material GaN is a binary compound, only the effect of the interface roughness needs to be taken into account in the calculation for the GaN/$Al_xGa_{1-x}N$ QWs.

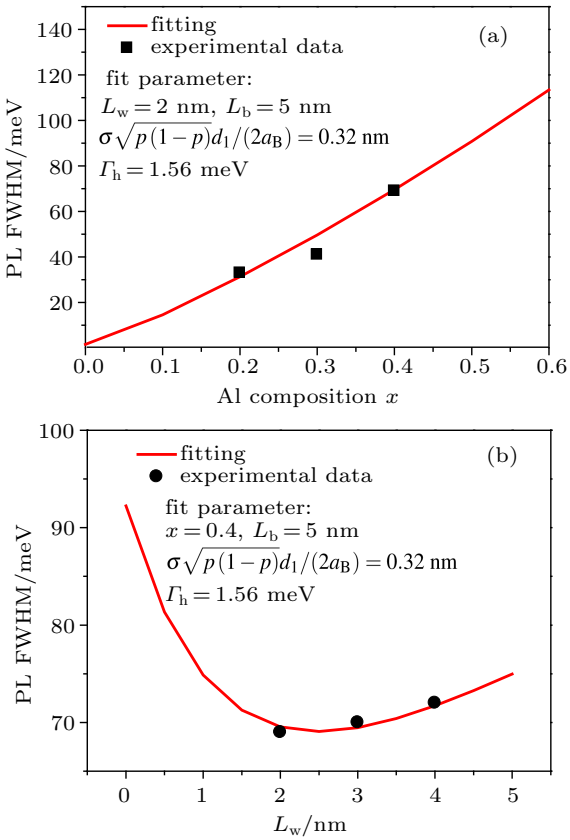


Fig. 2. (color online) PL FWHM of GaN/$Al_xGa_{1-x}N$ QW samples as a function of (a) the Al composition in the $Al_xGa_{1-x}N$ barrier and (b) the QW thickness. Solid squares are experimental results, and solid curves are theoretical fitting.

The schematic diagram of the band structure of one period of the GaN/AlGaN QWs is shown in Fig. 3. It can be seen clearly from the figure that due to the built-in field, the energy band tilts, in consequence, a triangle potential well forms in the GaN well layer. By assuming the energy levels of electrons in the conduction band and those of holes in the valence band for the GaN well, only the lowest energy levels of electrons and holes need to be considered. Then the transition energy of electrons from the conduction band to the valance band can be expressed as

$$E = E_{0\rm e} + E_{0\rm h} + E_{\rm g} - E_{\rm b} - eF_{\rm w}L_{\rm w}, \tag{2}$$

where $E_{0\rm e}$ and $E_{0\rm h}$ are the subband energies of electrons and holes in triangle potential well, respectively, $E_{\rm g}$ is the band gap of the GaN well, $E_{\rm b}$ is the bound energy of excitons, $F_{\rm w}$ is the field in the well, and $L_{\rm w}$ is the width of the well. The term $eF_{\rm w}L_{\rm w}$ represents the field-induced red shift of the gap.

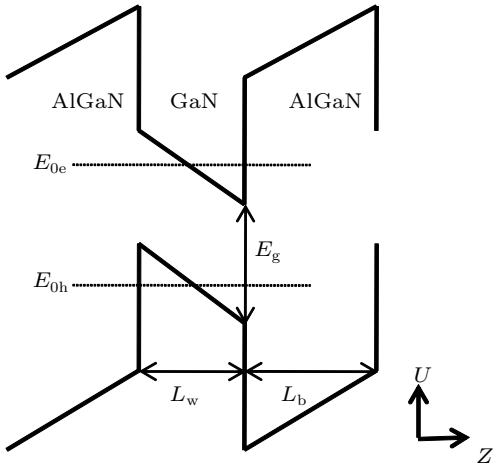


Fig. 3. Schematic graph of the band structure in GaN/AlGaN QWs, where L_w and L_b are the widths of the well and the barrier, respectively.

In the first order approximation, the variation of the emitted photon energy, δE, is given by the sum of the fluctuations in the band gap of the GaN well, the energies of electrons and holes, the bound energy of the excitons, and eF_wL_w

$$\delta E = \delta E_{0e} + \delta E_{0h} - \delta\left(eF_wL_w\right). \tag{3}$$

In the above equation, the variation of the bound energy of the excitons is neglected, which is reasonable when the well width is larger than the Bohr radius of the excitons.

In a triangle potential well, the ground subband energies of electrons and holes can be given as[19]

$$E_{0e/0h} = A_{0e/0h}F_w^{2/3}, \tag{4}$$

where

$$A_{0e/0h} = 2.338\left(\frac{\hbar^2e^2}{2m^*_{0e/0h}}\right)^{1/3},$$

here $\hbar$ is the reduced Planck constant, e is the electron charge, and $m^*_{0e/0h}$ is the effective mass of electron/hole.

As for the polarization field, according to the periodic boundary condition, the field in the well material of the QWs can be given as[14,20]

$$F_w = \frac{\left(P_b - P_w\right)L_b}{\varepsilon_wL_b + \varepsilon_bL_w}, \tag{5}$$

where L_w and L_b are the well and the barrier widths, respectively, $\varepsilon_w\left(\varepsilon_b\right)$ is the well (barrier) dielectric constant, and $P_w\left(P_b\right)$ is the macroscopic polarization.

For the GaN/$Al_xGa_{1-x}N$ QWs, we assume that the GaN well is strain-relaxed and the $Al_xGa_{1-x}N$ barrier pseudomorphically grows on the unstrained GaN layer, thus the $Al_xGa_{1-x}N$ barrier is under tensile strain, so

$$P_w = P_{sp}\left(\mathrm{GaN}\right), \tag{6}$$

$$P_b = P_{sp}\left(Al_xGa_{1-x}N\right) + P_{pE}\left(Al_xGa_{1-x}N\right), \tag{7}$$

where $P_{sp}\left(Al_xGa_{1-x}N\right)$ can be obtained by using the linear interpolation with the spontaneous polarizations of GaN and AlN. The piezoelectric polarization in the c-axis direction of wurtzite nitrides can be determined by[21]

$$P_{pE} = 2\frac{a\left(x\right) - a_0\left(x\right)}{a_0\left(x\right)}\left(e_{31}\left(x\right) - e_{33}\left(x\right)\frac{c_{13}\left(x\right)}{c_{33}\left(x\right)}\right), \tag{8}$$

where $e_{31}\left(x\right)$ and $e_{33}\left(x\right)$ are the piezoelectric coefficients, $c_{13}\left(x\right)$ and $c_{33}\left(x\right)$ are the elastic constants, and $a_0\left(x\right)$ is the equilibrium value of the lattice parameter. Substituting P_w and P_b from Eqs. (6)–(8) into Eq. (5), we can obtain the polarization field and then the energy band of electrons and holes in Eq. (4). Finally, the fluctuation of the energy band of electrons and holes and the third term in Eq. (3) can be obtained as follows:

$$\delta E_{0e} = \frac{\partial E_{0e}}{\partial F_w}\frac{\partial F_w}{\partial L_b}\delta L_b + \frac{\partial E_{0e}}{\partial F_w}\frac{\partial F_w}{\partial L_w}\delta L_w, \tag{9}$$

$$\delta E_{0h} = \frac{\partial E_{0h}}{\partial F_w}\frac{\partial F_w}{\partial L_b}\delta L_b + \frac{\partial E_{0h}}{\partial F_w}\frac{\partial F_w}{\partial L_w}\delta L_w, \tag{10}$$

$$\delta\left(eF_wL_w\right) = eL_w\left(\frac{\partial F_w}{\partial L_w}\delta L_w + \frac{\partial F_w}{\partial L_b}\delta L_b\right) + eF_w\delta L_w, \tag{11}$$

where δL_b and δL_w are the fluctuations of the widths of the barrier and the well, respectively. By applying Eqs. (4) and (5), we can obtain the differential expression as

$$\frac{\partial E_{0e}}{\partial F_w} = \frac{2A_{0e}}{3}F_w^{-1/3}, \tag{12}$$

$$\frac{\partial E_{0h}}{\partial F_w} = \frac{2A_{0h}}{3}F_w^{-1/3}, \tag{13}$$

$$\frac{\partial F_w}{\partial L_b} = \frac{\left(P_b - P_w\right)L_w\varepsilon_w}{\left(\varepsilon_wL_b + \varepsilon_bL_w\right)^2}, \tag{14}$$

$$\frac{\partial F_w}{\partial L_w} = \frac{\left(P_b - P_w\right)L_b\varepsilon_b}{\left(\varepsilon_wL_b + \varepsilon_bL_w\right)^2}, \tag{15}$$

then equations (9)–(11) can be expressed as

$$\delta E_{0e} = \frac{2}{3}A_{0e}F_w^{\frac{2}{3}}\frac{1}{\varepsilon_wL_b + \varepsilon_bL_w}\left(\frac{L_w\varepsilon_w}{L_b}\delta L_b - \varepsilon_b\delta L_w\right), \tag{16}$$

$$\delta E_{0h} = \frac{2}{3}A_{0h}F_w^{\frac{2}{3}}\frac{1}{\varepsilon_wL_b + \varepsilon_bL_w}\left(\frac{L_w\varepsilon_w}{L_b}\delta L_b - \varepsilon_b\delta L_w\right), \tag{17}$$

$$\delta\left(eF_wL_w\right) = eF_w\frac{\varepsilon_w}{\varepsilon_wL_b + \varepsilon_bL_w}\left(L_b\delta L_w + \frac{L_w^2}{L_b}\delta L_b\right). \tag{18}$$

Substituting Eqs. (16)–(18) into Eq. (3) and using a set of parameters,[22,23] we obtain the variation of the emitted photon energy δE.

Singh and Bajaj have developed a statistics model to describe the PL linewidth of a quantum well in terms of the interface roughness. In their opinion, the interfacial region is characterized by parameter σ representing the fluctuations in thickness along the growth direction and parameter d_1 representing the lateral average dimensions of the islands with coverage p, then the linewidth due to the interface roughness can be expressed as[11,24]

$$\Gamma = \left(\sigma\sqrt{p\left(1-p\right)}\frac{d_1}{2a_B}\right)\frac{\delta E}{\delta L_z}, \tag{19}$$

where a_B is the Bohr radius of the excitons, $\sigma\sqrt{p(1-p)}d_1/2a_B$ describes the standard fluctuation, $\delta E/\delta L_z$ represents the change in the excitonic emission energy due to the fluctuation of the width of the barrier or the well. For our case, using $\delta L_b = \delta L_w$, we can obtain $\delta E/\delta L_z$ by using δE to divide δL_b or δL_w in Eq. (3), and then substitute $\delta E_{imh}/\delta L_z$ into Eq. (19) to obtain Γ_{imh}. Next, considering the intrinsic linewidth Γ_h as a constant, we use the total linewidth (the sum of Γ_{imh} and Γ_h) to fit the experimental results. We find that the PL FWHM variations observed experimentally are well fit with the theoretical results as shown in Fig. 2, the fitting parameters are $\sigma\sqrt{p(1-p)}d_1/(2a_B) \sim 0.32$ nm and intrinsic linewidth $\Gamma_h \sim 1.56$ meV, which agree well with the previous reports.[25,26] The fitting curves show that the PL FWHM increases with the Al composition in the $Al_xGa_{1-x}N$ barrier. Whereas, as can be seen from Fig. 2(b), the variation in FWHM with the QW thickness is nonmonotonic, it decreases first and then increases a little, which is consistent with the CdTe QWs.[14] The experimental results in Fig. 2 can be qualitatively interpreted as follows. For Fig. 2(a), as the Al composition increases, the polarization in the barrier increases, resulting in the ramp of the built-up field in the QWs, the large field tends to cause fluctuation of the energy band. While for Fig. 2(b), when the well width is small, the polarization field hardly changes with the well width ($F_w \propto (1 + L_w/L_b)^{-1}$, L_w/L_b is small), then the main factor that influences the FWHM is $-eL_w\delta F_w$, so it decreases first with increasing well width. However, when the well width continues to increase, F_w would follow the variation of L_w, as a result, the FWHM increases with increasing L_w.

Note that the model is based on the triangle potential well, and electrons and holes are at the bottoms of the corresponding energy bands. Actually, in very narrow wells, the triangle potential well model is invalid. It is shown via calculation that for the barrier with Al composition $x = 0.4$ and a thickness of 5 nm, the model can be used when the well thickness is larger than 2 nm. What is more, other factors (such as the surface field and the field fluctuation due to active defects) that may affect the built-in field are neglectable, the main contribution should be the fluctuations of the width due to the interface roughness. Most important of all, the model output is very close to the experiment result.

4. Conclusion

We have investigated the optical FWHM of the GaN/$Al_xGa_{1-x}N$ QWs grown by MOCVD. It is experimentally found that the PL FWHM increases significantly with increasing Al composition in the barrier, while it increases slightly with the well width. We interpret the experimental results using a theoretical model, in which the interface roughness due to the fluctuations in both the well and the barrier widths is taken into account as the main factor determining the PL FWHM. Although the real interface is likely to be more complicated than that in our model, our model does describe the main features of the interface and captures the physics of the problem.

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