The exciton-longitudinal-optical-phonon coupling in InGaN/GaN single quantum wells with various cap layer thicknesses*

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(Received 28 February 2010; revised manuscript received 13 May 2010)

This paper studies the exciton–longitudinal-optical-phonon coupling in InGaN/GaN single quantum wells with various cap layer thicknesses by low temperature photoluminescence (PL) measurements. With increasing cap layer thickness, the PL peak energy shifts to lower energy and the coupling strength between the exciton and longitudinal-optical (LO) phonon, described by Huang–Rhys factor, increases remarkably due to an enhancement of the internal electric field. With increasing excitation intensity, the zero-phonon peak shows a blueshift and the Huang–Rhys factor decreases. These results reveal that there is a large built-in electric field in the well layer and the exciton–LO-phonon coupling is strongly affected by the thickness of the cap layer.

Keywords: exciton–longitudinal-optical-phonon, InGaN/GaN single quantum well, GaN cap layer, Huang–Rhys factor

PACC: 7865K, 7855E, 6320K

1. Introduction

GaN-based quantum well (QW) and heterostructures have attracted intensive research due to their application in light emitting devices operating from the ultraviolet to the entire visible spectra region[1] and in electronic devices such as field-effect transistors.[2] Such devices are highly demanded in high-density data storage, higher efficiency lighting sources, finer scale laser printer and full colour display.[3] It is known that strong internal electric field is intrinsically present in wurtzite GaN-based heterostructures due to which result in the quantum confined Stark effect (QCSE) in the InGaN QWs.[4,5] For the GaN-based QWs, their optical and electronic properties are strongly affected by the interaction between excitons and longitudinal-optical (LO) phonons[6−8] which in turn plays an important role in determining the performance of GaN-based electronic and optoelectronic devices. It has been shown that as the indium fractions or the well widths increase, the interaction between excitons and phonons increases steadily in single InGaN/GaN QWs[8,9] due to an increase of the electron–hole separation originated from the existence of built-in electric field. This electron–hole separation was caused by strain-enhanced built-in electric field in Ref. [8] and by increased well width in Ref. [9], respectively. While in both cases, the cap layer thickness is kept as a constant for different samples. In this paper, exciton–LO phonon interaction in InGaN/GaN single QWs with varying cap layer thicknesses is studied. It is found that the exciton–LO phonon interaction is strongly dependent on the thickness of the top GaN cap layer. The larger the cap layer thickness is, the stronger the interaction is. The excitation-intensity-dependent photoluminescence (PL) spectra demonstrate the existence of internal electric field induced by spontaneous and piezoelectric polarisations. The results demonstrate that the built-in electric field is also strongly dependent on the cap layer thickness.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 60876007 and 10974165) and the Research Program of Xiamen Municipal Science and Technology Bureau, China (Grant No. 2006AA03Z110).

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2. Experiment

Four InGaN/GaN single QW samples studied in this paper were grown by a metal organic chemical vapour deposition system on 5.08 cm. diameter (0001)-orientated sapphire substrates as follows: a 30 nm GaN nucleation layer, a 2.0 μm thick undoped GaN buffer layer, a single QW consisting of a 3-nm-thick InGaN well layer with 10% nominal indium concentration and GaN cap layer with the thicknesses of 3, 5, 10, and 15 nm. The single QW structures are schematically shown in Fig. 1. The PL emission of the single QW samples were confirmed under excitation by using 325-nm He–Cd laser with a spot size of about 150 μm in diameter at a sample temperature of 15 K. The excitation-intensity-dependent PL spectra were performed by using frequency-tripled Nd: YAG laser emitting at 355 nm with a pulse-width of 5 ns and a repetition rate of 50 Hz.

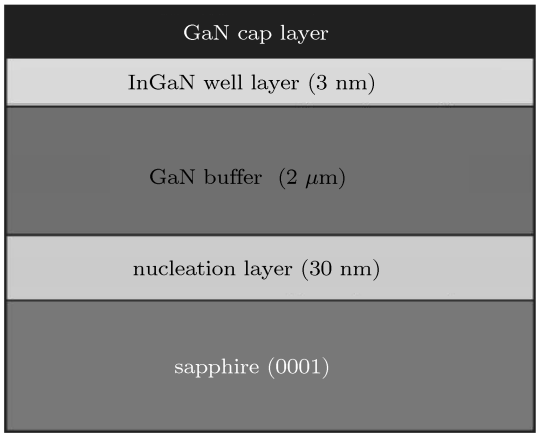


Fig. 1. Schematic diagram of the single QW. The cap layer thicknesses are 3, 5, 10 and 15 nm.

3. Result and discussion

One of the important features of exciton–phonon interaction is the appearance of phonon replicas of excitons in the PL spectra.[7,8] The intensities of these phonon replicas depend strongly on the excition-phonon coupling strength. Within the Frank–Condon approximation, the distribution of emission intensities between the nth-order phonon replica (I_n) and the zero-phonon peak (I_0) can be described in terms of the Huang–Rhys factor S, which is expressed by[10]

$$I_n = I_0 \frac{S^n}{n!}, \quad n = 0, 1, 2, \ldots. \tag{1}$$

Here, n is the number of LO phonons involved.

The low temperature (15 K) PL spectra of the four single QW samples were shown in Fig. 2 with the excitation of a 325 nm He–Cd laser. The PL spectra were normalised so that the strongest peaks were of the same intensity. The spectra of these samples exhibited a main peak and two phonon replicas at the lower energy side which were assigned to the recombination assisted by emitting one and two LO phonons, respectively. With increasing cap layer thickness, the zero-phonon peak shifted from 3.268 eV for a cap layer thickness of 3 nm to 3.154 eV for a cap layer thickness of 15 nm. Similar behaviour of the energy shift has also been reported previously. Leroux *et al.*[11] reported a blueshift with decreasing the barrier thickness in the AlGaN/GaN multiple QWs. This was attributed to the strong built-in electric field induced by the difference in spontaneous polarisation between the well and barrier layers. Similarly, with the increase of barrier thickness, Yu *et al.*[12] reported a redshift of PL peak energy of InGaN/GaN multiple QWs due to an enhancement of the QCSE induced by the spontaneous and piezoelectric field. An important feature of InGaN/GaN QW structures is that a strong built-in electric field exists along the grown axis inside the well layer caused by spontaneous and piezoelectric polarisations. Therefore, the redshift of the main peak with increasing cap layer thickness can be understood as the effect of enhanced QCSE caused by the built-in electric field. Because the well width is same for all samples, our results indicate that a stronger built-in electric field is expected for larger cap layer thickness. Apart from the peak shift, it was also observed that the intensity of the phonon replicas increases gradually with increasing cap layer thickness, as shown in Fig. 2. The energy spacing between the adjacent peaks is about 90 meV, corresponding to the LO phonon energy in InGaN/GaN QWs.[6,8,13] The Huang–Rhys factor can be estimated from the distribution of the emission intensities between the LO phonon replicas and the zero-phonon peak by Eq. (1). However, the zero-phonon peak contains contributions from all the recombining excitons, whereas only those that were strongly localised contribute significantly to the phonon replicas. [6,9,13,14] Therefore, the comparison between the intensities of the two LO phonon replicas is more accurate than the comparison between the first LO and the zero-phonon peak. Equation (1) can be transformed into[13]

$$S_n = (n+1)\frac{I_n + 1}{I_n}, \quad n = 0, 1, 2, \ldots. \tag{2}$$

Thus the $S_1(= 2I_2/I_1)$ describes the strength ratio between the 2LO phonon replicas and the 1LO phonon replicas and $S_0(= I_1/I_0)$ describes the strength ratio between the LO phonon replicas and the zero-phonon peak. The PL spectrum was fitted by using Gaussian functions with subtraction of a linear background. The energy spacing of 90 meV between the adjacent peaks was taken as a fitting parameter, while the full width at half maximum (FWHM) was taken the same for all the peaks. By this procedure, we obtained the areas of the two LO phonon replicas from which S_1 was estimated.

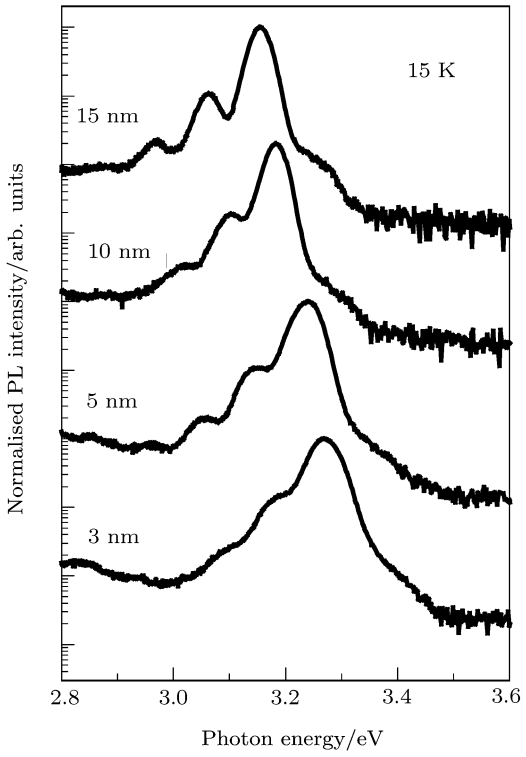


Fig. 2. Normalised PL spectra at 15 K of the InGaN/GaN single QWs with different cap layer thicknesses on a semilogarithmic scale. The spectra were shifted vertically for clarity.

Figure 3 shows the fitted result for the 15 K PL spectrum of the sample with a cap layer thickness of 15 nm. Applying this method to other samples, we obtained S_1 as a function of the cap layer thickness given in Fig. 4. It is obvious that, with increasing cap layer thickness, S_1 increases steadily, indicating an increase of the coupling strength between the excitons and LO phonons.

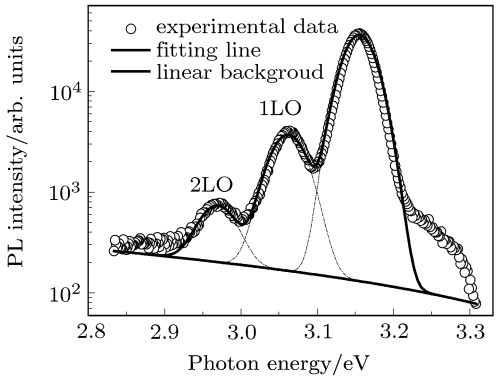


Fig. 3. The 15 K PL spectra of the InGaN/GaN single QW with a cap layer thickness of 15 nm. The lines show the Gaussian fitting and linear background.

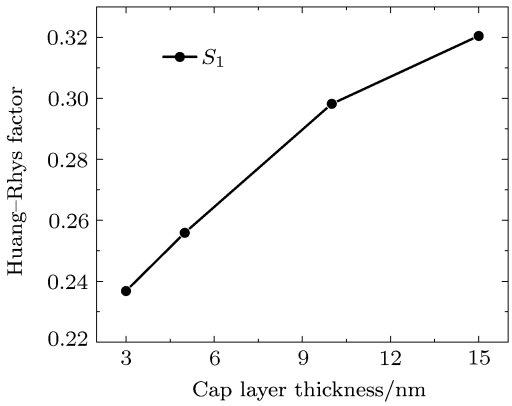


Fig. 4. Huang–Rhys factor as a function of the cap layer thickness.

Theoretical descriptions of the exciton–LO-phonon interaction based on the adiabatic approximation indicate that the Huang–Rhys factor S depends strongly on the spatial distributions of the electron and hole charge densities.[7,15] Generally, the Huang–Rhys factor S becomes larger as the distance between the electrons and the holes increases due to the reduced overlap of electron and hole charge densities.[6,7] As described above, a strong built-in electric field exists along the grown axis in InGaN/GaN QW structures. Such an electric field pushes the electrons and the holes to opposite sides of the well, leading to an increase of distance between the electron and the hole. In our special single QW samples where the well width is constant, the larger S value can only be attributed to the enhanced internal electric field for a larger cap layer thickness. This is consistent with the result observed for the redshift of the emission peak discussed above. The behaviors of both the emission peak and the S value demonstrate that the built-in electric field is strongly dependent on the cap layer thickness, and becomes stronger for larger cap layer thicknesses.

In order to confirm the above explanation, the excitation-intensity-dependent PL spectra for the sample with a cap layer thickness of 15 nm were measured by using the YAG pulse laser at 15 K, as shown in Fig. 5. The excitation intensity was varied from 8 nJ to 2 μJ. Under a lower excitation intensity, a dominant peak with LO phonon replicas was observed. With increasing excitation intensity, the LO phonon replicas disappear gradually and the zero-phonon peak exhibits a blueshift. From the curves a–c in Fig. 5 we can see that the zero-phonon peak shifts to the high energy with the increase of excitation intensity, which can be understood as a result of the screening of the internal electric field.[16] When the internal electric field was gradually screened, the tilt of energy band was suppressed and the effective band gap was increased gradually, resulting in the blueshift. From the curves d to e in Fig. 5, when the excitation intensity is high enough, the internal electric field was fully screened and the line broadening can be explained by the band-filling effect of the localised states at potential fluctuations.

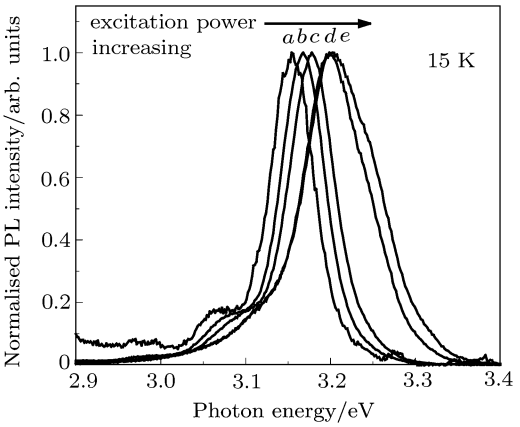


Fig. 5. Normalised PL spectra of the InGaN/GaN single QW with a cap layer thickness of 15 nm as the excitation intensity from 8 nJ to 2 μJ from curves a to e.

The Huang–Rhys factor S and the zero-phonon peak energy as a function of the excitation intensity were shown in Fig. 6. When the excitation intensity is high enough the LO phonon replicas are no longer resolved. Similar to the energy shift, the decreased S value can be explained by considering the variation of internal electric field. As discussed above, the distance along the growth axis between electrons and holes was determined by the internal electric field. With increasing excitation intensity, the internal electric field is gradually screened and the distance between the electron and hole charge distributions becomes shorter which leads to the reduction of the S value. Such a result is consistent with the work of Graham *et al.*[8] in which the reduction of the excitation density resulted in a redshift of the PL spectrum and an increase of S value.

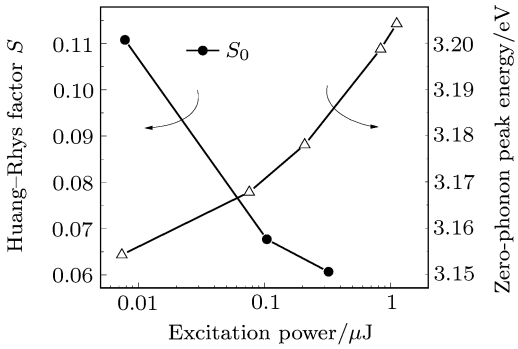


Fig. 6. Huang–Rhys factor and the zero-phonon peak energy obtained from Fig. 5 as a function of the excitation intensity.

4. Summary

In conclusion, we have studied the low temperature PL spectrum of excitons and LO phonons in InGaN/GaN single QW samples as a function of GaN cap layer thickness. With increasing cap layer thickness, the zero-phonon peak energy decreased and the Huang–Rhys factor increased. The excitation-intensity-dependent PL measurement revealed a blueshift of the zero-phonon peak and a decrease of the Huang–Rhys factor with increasing excitation intensity. These results obviously demonstrate the existence of the internal electric field, and the larger the cap layer thickness is, the stronger the electric field is. Consequently, the thickness of GaN cap layer, which strongly modifies the internal electric field, plays a crucial role in determining the electronic and optical properties of the devices based on such single QW or heterostructures.

References

[1] Mukai T, Yamada M and Nakamura S 1999 *Jpn. J. Appl. Phys.* **38** 3976

[2] Wu Y F, Keller B P, Fini P, Keller S, Jenkins T J, Kehias L T, Denbaars S P and Mishra U K 1998 *IEEE Electr. Device Lett.* **19** 50

[3] Nakamura S, Senoh M, Nagahama S, Iwasa N, Matsushita T and Mukai T 2000 *Appl. Phys. Lett.* **76** 22
[4] Fiorentini V, Bernardini F, Della Sala F, Di Carlo A and Lugli P 1999 *Phys. Rev.* B **60** 8849
[5] Tao R C, Yu T J, Jia C Y, Chen Z Z, Qin Z X and Zhang G Y 2009 *Chin. Phys.* B **18** 2603
[6] Zhang X B, Taliercio T, Kolliakos S and Lefebvre P 2001 *J. Phys.-Condens. Mat.* **13** 7053.
[7] Kalliakos S, Zhang X B, Taliercio T, Lefebvre P, Gil B, Grandjean N, Damilano B and Massies J 2002 *Appl. Phys. Lett.* **80** 428
[8] Graham D M, Soltani-Vala A, Dawson P, Godfrey M J, Smeeton T M, Barnard J S, Kappers M J, Humphreys C J and Thrush E J 2005 *J. Appl. Phys.* **97** 103508
[9] Olaizola S M, Fan W H, Mowbray D J, Skolnick M S, Parbrook P J and Fox A M 2007 *Superlattice Microst.* **41** 419
[10] Smith M, Lin J Y, Jiang H X, Khan A, Chen Q, Salvador A, Botchkarev A, Kim W and Morkoc H 1997 *Appl. Phys. Lett.* **70** 2882
[11] Leroux M, Grandjean N, Massies J, Gil B, Lefebvre P and Bigenwald P 1999 *Phys. Rev.* B **60** 1496
[12] Yu Y J, Ryu M Y, Yu P W, Kim D J and Park S J 2001 *J. Korean Phys. Soc.* **38** 134
[13] Pecharroman-Gallego R, Edwards P R, Martin R W and Watson I M 2002 *Mat. Sci. Eng. B-Solid* **93** 94
[14] Paskov P P, Holtz P O, Monemar B, Kamiyama S, Iwaya M, Amano H and Akasaki I 2002 *Phys. Status Solidi* B **234** 755
[15] Heitz R, Mukhametzhanov I, Stier O, Madhukar A and Bimberg D 1999 *Phys. Rev. Lett.* **83** 4654
[16] Kuokstis E, Yang J W, Simin G, Khan M A, Gaska R and Shur M S 2002 *Appl. Phys. Lett.* **80** 977