Exciton–phonon interaction in $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ multiple quantum wells*

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The exciton–phonon interaction in $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ multiple quantum wells (MQWs) is studied by deep-ultraviolet time-integrated and time-resolved photoluminescence (PL). Up to four longitudinal-optical (LO) phonon replicas of exciton recombination are observed, indicating the strong coupling of excitons with LO phonons in the MQWs. Moreover, the exciton–phonon coupling strength in the MQWs is quantified by the Huang–Rhys factor, and it keeps almost constant in a temperature range from 10 K to 120 K. This result can be explained in terms of effects of fluctuations in the well thickness in the MQWs and the temperature on the exciton–phonon interaction.

Keywords: exciton–phonon coupling, AlGaN quantum wells, deep-ultraviolet photoluminescence

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1. Introduction

AlGaN-based multiple quantum wells (MQWs) with high Al content have attracted intensive attention for their device applications in deep-ultraviolet (DUV) light emitting diodes (LEDs),[1–3] laser diodes (LDs),[4,5] and detectors.[6,7] Understanding light emission and absorption mechanisms in AlGaN MQWs is important for developing these devices. The exciton–phonon interaction, one of the intrinsic physical properties, is an important effect that appears in the optical process. The lattice vibration perturbs the motions of electrons/excitons in different ways, resulting in the various types of electron/exciton–phonon interactions, such as the Fröhlich interaction with longitudinal optical (LO) phonons, deformation–potential interactions with optical and acoustic phonons and piezoelectric interaction with acoustic phonons. The Fröhlich interaction is stronger than the other interactions due to the ionic nature of III–nitrides, which has a strong influence on the optical and transport properties of III–nitrides.[8,9]

Previous researches on exciton–phonon interactions in GaN epilayers,[10] AlN epilayers,[9] InGaN/GaN MQWs, and GaN/AlGaN MQWs[11] have been reported. The LO phonons in GaN and AlN epilayers have energies of about 92 meV[10,12] and 110 meV,[9] respectively. The exciton–phonon coupling strengths in various epitaxial structures, such as thin films, MQWs and quantum dots determine the optical transitions. As far as we know, the exciton–phonon interactions in the AlGaN/AlGaN MQWs have been less studied due to the lack of high quality materials. Additionally, the experimental results as reported on GaN-based quantum wells such as InGaN/GaN and GaN/AlGaN MQWs are controversial because of the complicated exciton–phonon interactions in these low dimensional GaN-based structures. Both reduced and enhanced exciton–phonon interactions in the nitride MQWs in comparison with those in nitride bulk materials have been reported.[10,11,13]

In this paper, deep-ultraviolet time-integrated and time-resolved photoluminescence (PL) measurements are performed on the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs fabricated by using metal–organic chemical vapor deposition (MOCVD). Up to four LO-phonon replicas of exciton recombination are observed. The Huang–Rhys factor, which reflects the exciton–phonon coupling strength, is deduced from the ratio of the emission intensity of phonon replicas to that of the zero phonon line. The temperature insensitivity of the Huang–Rhys factor is explained in terms of the effects of fluctuations in the well thickness and the temperature on the exciton–phonon interaction in the MQWs.

2. Experiments

The MQWs used in this study were grown on a *c*-plane sapphire substrate by metal organic chemical vapor deposition. Prior to the growth of AlGaN/AlGaN MQWs, an AlN buffer layer about 600 nm was grown on the sapphire substrate, which was followed by an $Al_{0.62}Ga_{0.38}N$ epilayer with

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a thickness of about 600 nm. Then the AlGaN/AlGaN MQWs of 20 periods were deposited on the epilayer. The Al compositions in the AlGaN wells and AlGaN barriers were 0.4 and 0.53, respectively, while the thickness values of AlGaN wells and AlGaN barriers were 2.3 nm and 7.3 nm, respectively. The Al compositions and the thickness values of AlGaN wells and AlGaN barriers were determined by the measurement of XRD ω scans of (002) planes. Finally, an $Al_{0.53}Ga_{0.47}N$ cap layer about 8 nm in thickness was covered on the MQWs.

The time-integrated and time-resolved PL measurements were performed by using our home-made deep ultraviolet laser spectroscopy system with a frequency-tripled fs Ti:sapphire laser as the excitation source, providing an excitation wavelength of about 237 nm. The diameter of the excitation area on the sample was about 100 μm, and the sample temperature was varied from 10 K to 180 K. Time-resolved PL spectra were recorded by a stand streak camera acquisition system, and the overall time resolution of the system is less than 16 ps. For the time-integrated PL, the spectral resolution was about 0.03 nm at 300 nm.

3. Results and discussion

The time-integrated photoluminescence spectrum of $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs measured at $T = 10$ K is plotted in Fig. 1(a). Five transition peaks at 4.591, 4.493, 4.395, 4.297, and 4.199 eV with the values of full width at half maximum (FWHM) of 62, 70, 70, 70, and 70 meV, denoted as A, A_1, A_2, A_3, and A_4, respectively, are clearly resolved in the PL spectrum by a multi-peak Gaussian fitting. They originate from the $Al_{0.4}Ga_{0.6}N$ well region. Figure 1(b) shows the temperature-dependent PL spectra of the MQWs obtained from 10 K to 180 K. The solid circles indicate the main peak positions of the spectra. In Fig. 1(b), the main emission peaks of A in the PL spectra exhibit a redshift of 17 meV with temperature increasing from 10 K to 120 K, and then followed by a blueshift of 18 meV with increasing temperature from 120 K to 180 K, which is a common characteristic of localized excitons in III–nitride alloy material, resulting from the potential fluctuations.[14–16] It has been reported that the fluctuations in alloy content and well width largely determine the PL FWHM.[17] So the broad PL FWHM further confirms the existence of localized excitons in the MQWs. Thus, the main emission peaks of A as shown in Fig. 1(b) are assigned to be the localized exciton emissions. Furthermore, it is interesting to note that the energy separation between these successive lines in Fig. 1(a) is about 98 meV. LO phonon replicas have been observed in GaN epilayers and AlN epilayers with LO phonon energies of 92 meV[10,12] and 110 meV,[9] respectively. The energy separation of 98 meV in the PL spectrum is between the two LO phonon energies in GaN and AlN. It is more likely to be the LO phonon energy in the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs. Therefore, the four emission peaks of A_1, A_2, A_3, and A_4 should be assigned to be four phonon replicas of the localized exciton emission. The experimental observations have been reported of two and four phonon replicas in the PL spectrum from InGaN/GaN MQWs[11,18] and two phonon replicas in GaN/AlGaN MQWs.[11,19] As far as we known, the experimental observations of phonon replicas in the PL spectra from AlGaN/AlGaN MQWs have not been reported so far in the literature. In addition, it is worth noting that the phonon replica lines appearing in the PL spectra are an indicator of a strong exciton–phonon interaction in the sample. It is also noted that no other type of phonon replica lines are observed in the PL spectra except LO phonon lines, which indicates that the exciton-LO phonon interaction is the strongest in the MQWs.

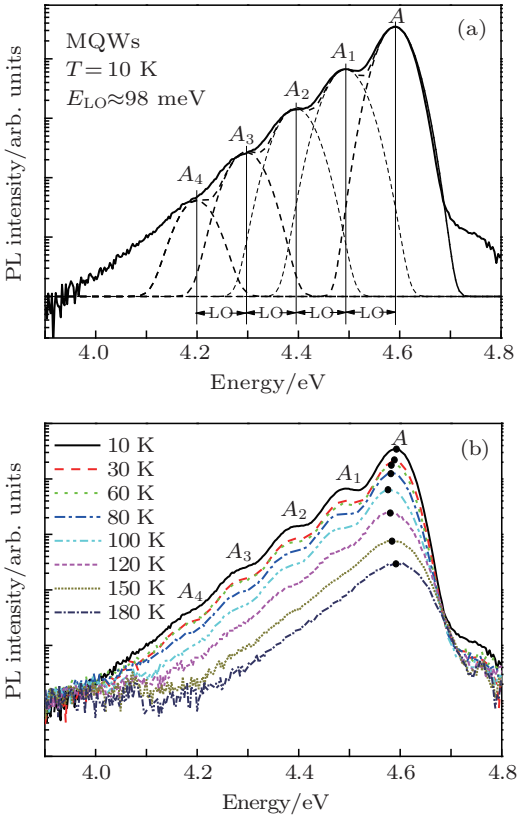


Fig. 1. (color online) (a) PL spectrum of $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs at $T = 10$ K, showing four phonon replicas of the excitonic transition at 4.591 eV with an LO phonon energy of about 98 meV by a multi-peak Gaussian fitting. The dotted lines are the fitting curves. (b) Temperature dependent PL spectra of $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs. The solid circles indicate the main peak positions of the spectra.

In order to investigate the recombination properties of the main emission peak and the phonon replicas in the PL spectrum of the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs, the time-resolved PL measurements are performed. Figure 2 shows the decay signals for the main emission A and its phonon replicas A_1, A_2, and A_3 at 10 K. Each emission peak exhibits a single exponential decay curve, and the recombination lifetimes of A,

A_1, A_2, and A_3 are about 4.8 ns, 5.8 ns, 5.8 ns, and 5.6 ns, respectively. As reported in Ref. [3], the exciton recombination lifetimes of emission peaks in our sample are much longer than the exciton recombination lifetime of 600 ps in AlGaN/GaN MQWs at 5 K. The long lifetimes of the PL emission peaks in Fig. 2 may be due to the involvement of excitons localized by alloy disorder and well width fluctuation in the MQWs: the excitons have low probability to reach the nonradiative recombination centers and thus have long lifetimes.[20] In addition, it should be noted that the recombination lifetime of the main emission peak A is shorter than the lifetimes of the phonon replicas. In general, it is better to measure the lifetimes of LO phonon replicas instead of the non-phonon line to determine the PL lifetime in the bulk region, because the energies of LO phonon replicas are lower than the bandgap, and they can come out of the bulk, where any possible effect due to surface nonradiative recombination can be excluded.[21]

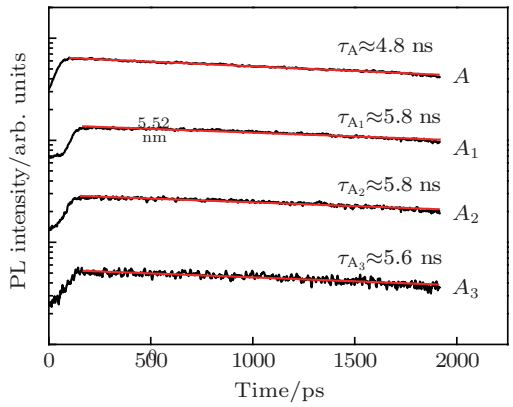


Fig. 2. (color online) PL decay curves for the main emission A and its phonon replicas A_1, A_2, and A_3 of the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs at 10 K. The recombination lifetimes of the emission peaks of A, A_1, A_2, and A_3 are about 4.8 ns, 5.8 ns, 5.8 ns, and 5.6 ns, respectively.

The distributions of the emission intensities among the main peak ($n = 0$) and the phonon replicas ($n = 1, 2, 3, 4$) depend strongly on the exciton–phonon coupling strength.[22,23] This coupling is expressed by the Huang–Rhys factor S within the Frank–Condon approximation. At low temperature, the relationship between the intensity of the n-th phonon replica I_n and the intensity of the main emission I_0 is given by[8]

$$I_n = I_0 \frac{S^n}{n!}, \tag{1}$$

where $n = 0, 1, 2, 3, \ldots$, with n representing the number of LO phonons involved.

Figure 3 shows I_n/I_0 as a function of n measured at 10 K for the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs. To extract the Huang–Rhys factor S, the observed relative intensities I_n/I_0 in Fig. 3 are fitted into Eq. (1). The S-parameter for the MQWs at 10 K obtained is $S = 0.23$, which is much larger than 0.0077 in bulk GaN reported in Ref. [8]. The large S value is due to the fact that the exciton–phonon interaction in III–nitride MQWs is enhanced by the symmetry properties of MQWs and alloy disorder.[11] However, it is also interesting to note that the value of S factor obtained from the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs is smaller than the S value of 0.556 obtained from $Al_{0.07}Ga_{0.93}N$ barriers in GaN/AlGaN MQW.[11] It may be due to the existence of impurities in the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs sample. Impurities or related defects in the MQWs may enhance the scattering of exciton–polariton, which may be conducible to momentum transfer, resulting in increasing the possibility of recombination without the participation of phonons. Then the number of phonons involved is reduced, therefore the coupling strength is weakened.[9]

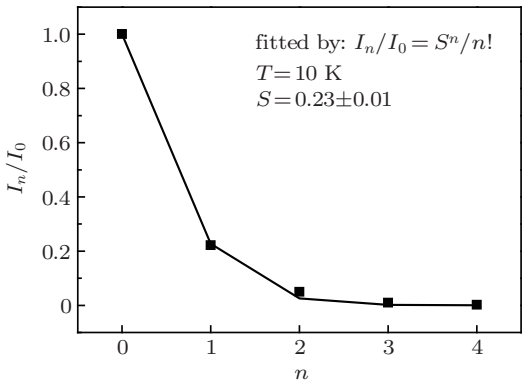


Fig. 3. Ratio of the PL emission intensity of n-th order phonon replicas I_n to that of the zero phonon line I_0, I_n/I_0, versus n at 10 K for the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs. The ratio I_n/I_0 is fitted to $I_n = I_0 S^n/n!$ from which the Huang–Rhys factor S can be obtained.

The Huang–Rhys factor in the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs is extracted by fitting the observed relative intensities I_n/I_0 to Eq. (1) in the temperature range from 10 K to 120 K. The temperature dependence of the Huang–Rhys factor S is demonstrated in Fig. 4. It can be seen from the figure that the parameter S is nearly constant with a mean value of 0.22 from 10 K to 120 K. In general, exciton–phonon coupling strength in a low-dimensional structure depends strongly on the distributions of the densities of electron and hole charges due to the effects of quantum confinement and internal electric field.[24–26] The reduced overlap of electron and hole charge densities in the low-dimensional structure makes the exciton–phonon coupling strength stronger. There is a strong internal electric field in quantum wells, caused by both piezoelectric and spontaneous polarization, which has considerable influence on the progressive separation of electron and hole charge densities with the increase in well width, resulting in a larger coupling constant S in the quantum wells. In addition, temperature has a great influence on the phonon-assisted recombination process in the III–nitride material.[27,28] It has been reported that the phonon-assisted excitonic transition probability in the III–nitride material depends on the kinetic energy of the excitons, so it is expected that the intensity ratio I_n/I_0

continuously increases with the increase in temperature.[8] As discussed above, the exciton–phonon coupling constant S depends on both the well width fluctuation and the temperature in the low-dimensional structure of III–nitride material. In our $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs, the nearly constant value of Huang–Rhys factor S may come from the contributions of the effects of the well width fluctuation and the temperature. A thermally enhanced phonon assisted transition probability with temperature increasing results in the increase of Huang–Rhys factor S in the MQWs. Meanwhile, a thermally activated transfer of excitons from a thick region to a relatively thin region in wells leads to the reduction of Huang–Rhys factor S. Eventually, a balance between the two contributions issues in the nearly invariable Huang–Rhys factor S in the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs.

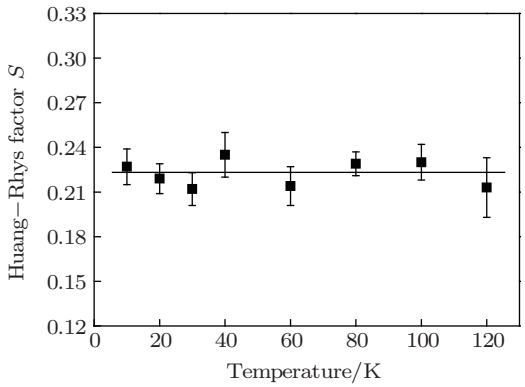


Fig. 4. Huang–Rhys factor S versus temperature for the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs. The solid line gives the Huang–Rhys factor S with a mean value of 0.22 from 10 K to 120 K.

4. Conclusions

In this work, the exciton–phonon interaction in the $Al_{0.4}Ga_{0.6}N/Al_{0.53}Ga_{0.47}N$ MQWs is investigated by deep-ultraviolet time-integrated and time-resolved PL. LO phonon replicas of exciton recombination with up to four phonons in the PL spectra are experimentally observed, indicating the strong coupling of excitons with LO phonons in our sample. Measuring the lifetimes of LO phonon replicas may be preferable to determine the PL recombination lifetime of the MQWs. In addition, it is found that the exciton–phonon coupling strength S is nearly invariable in the temperature range from 10 K to 120 K, which may be due to the fact that the balance between a thermally enhanced phonon assisted transition probability with temperature resulting in the increase of S factor and a thermally activated transfer of excitons from the thick region to the relatively thin region in wells leads to the reduction of S factor.

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