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Exciton-phonon interaction in InGaN/GaN and GaN/AlGaN multiple quantum wells

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The exciton-phonon coupling has been studied in $In_xGa_{1-x}N/GaN$ and $GaN/Al_xGa_{1-x}N$ multiple quantum wells (MQWs) and compared with that in $In_xGa_{1-x}N$ and GaN epilayers. Phonon replicas with up to four phonons can be well resolved only in the alloy regions of the MQWs ($In_xGa_{1-x}N$ or $Al_xGa_{1-x}N$) and was independent of the structure (well or barrier), while no phonon replicas of the exciton transitions were observed for the free-exciton transitions in the GaN and the localized exciton transitions in the $In_xGa_{1-x}N$ epilayers. It thus suggests that the symmetry properties of MQWs, which modifies the phonon dispersion relation, together with alloy disorder are responsible for the enhanced exciton-phonon interaction in III-nitride MQW. The coupling constant S of the exciton-phonon interaction is extracted for an $In_xGa_{1-x}N/GaN$ and $GaN/Al_xGa_{1-x}N$ MQW, and is found to be $S=0.802$ and 0.556, respectively. The implications of the modified exciton-phonon coupling in MQWs in terms of understanding the fundamental physics of this system as well as practical device applications are discussed. © *1997 American Institute of Physics.* [S0003-6951(97)03821-7]

The fabrication of GaN based devices such as blue-UV lasers and light emitting diodes (LEDs), solar-blind UV detectors, and high-power electronics have fueled recent intensive research in these materials.[1] Most optoelectronic devices based on III nitrides are designed using quantum well (QW) structures, InGaN/GaN or GaN/AlGaN, thus an understanding of the basic physical properties of III-nitride QWs is especially important. Recent work on the optical properties of III-nitride alloy systems and QWs has shown that localized exciton transitions dominate in these systems at low temperatures.[2–4] The interaction of the carrier or excitons with the phonons in III-nitride materials is of interest due to the large longitudinal optical (LO) phonon energies of InN (86 meV) and GaN (91 meV).[5] The strength of this interaction in various structures may determine the optical transitions in these structures. It may also determine the laser mechanisms in $In_xGa_{1-x}N/GaN$ multiple quantum well (MQW) blue lasers. However, this coupling strength has not been studied previously in III-nitride systems.

When there is a strong exciton-phonon or carrier-phonon interaction, the photoluminescence (PL) spectrum is characterized by a main emission line and several phonon replicas. The emitted photon energy $h\nu$ is given by

$$h\nu=E_0-nE_p, \quad n=0,1,2,\ldots, \tag{1}$$

where E_0 is the energy of the main emission peak, n indicates the number of phonons involved, and E_p is the phonon energy involved. Within a Franck–Condon model,[6,7] the coupling of the exciton transitions to the LO phonon is expressed by the exciton-phonon coupling constant, S. The intensity distribution of the phonon replicas is determined by S. For the case of relatively strong coupling, the emission intensity of the nth phonon replica $I_n(\gamma)$ and the principle emission line (I_0) is related by[6,7]

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

In this way, inspection of the emission spectra, $I_n(\gamma)$, for $n=0,1,2...$, gives information on the carrier or exciton-phonon coupling constant S.

The wurtzite GaN epilayer, $In_xGa_{1-x}N$ epilayer ($x\approx0.12$), and $In_xGa_{1-x}N/GaN$ MQW ($x\approx0.15$) samples studied here were grown by low pressure metal-organic chemical vapor deposition (MOCVD). The MQW sample is composed of 20 periods of alternating 45 Å $In_xGa_{1-x}N$ and 45 Å GaN barriers. Prior to the deposition of the GaN and $In_xGa_{1-x}N$ layers, a 50 nm GaN buffer layer was grown on a sapphire (Al_2O_3) substrate. The wurtzite $GaN/Al_xGa_{1-x}N$ MQW ($x\approx0.07$) sample was grown by reactive molecular beam epitaxy (MBE) on a sapphire (Al_2O_3) substrate with a 50 nm AlN buffer layer. The MQW is composed of 10 periods of alternating 25 Å GaN wells and 50 Å $Al_xGa_{1-x}N$ barriers. All of the samples are nominally undoped. The low temperature PL spectra were measured using a picosecond laser spectroscopy system.[2,3]

In Fig. 1, we have plotted the continuous-wave (cw) PL spectra of (a) the MBE grown $GaN/Al_xGa_{1-x}N$ MQW sample, (b) the MOCVD grown GaN epilayer, (c) the MOCVD grown $In_xGa_{1-x}N/GaN$ MQW, and (d) the MOCVD grown $In_xGa_{1-x}N$ epilayer. In Fig. 1(a), the $GaN/Al_xGa_{1-x}N$ MQW shows a dominant transition at 3.541 eV from the GaN well region, as well as a transition at 3.692 eV from the $Al_xGa_{1-x}N$ barrier region. The well transition at 3.541 eV, due to excitons localized by interface roughness in the MQW,[3] is blue shifted by 56 meV com-

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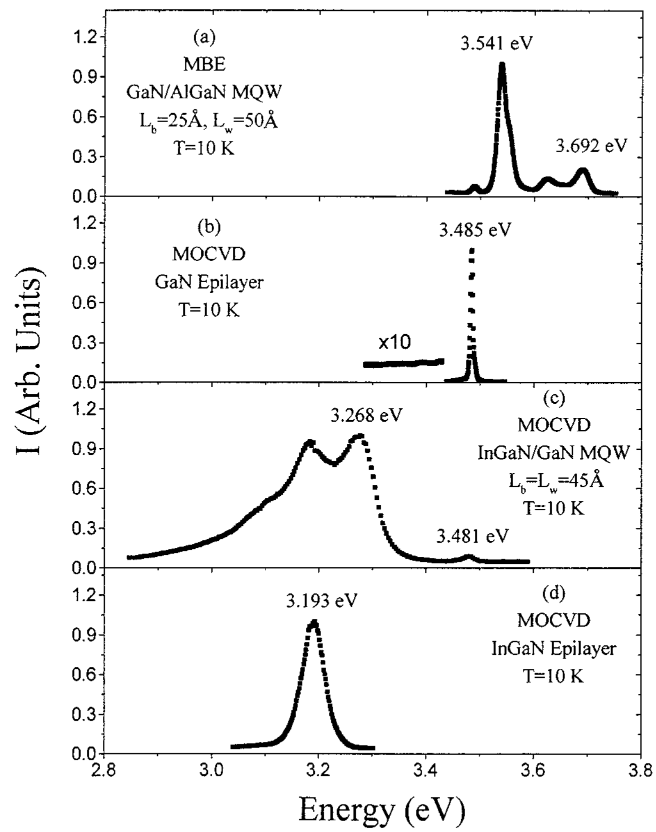

FIG. 1. cw PL spectra of (a) a GaN/$Al_xGa_{1-x}N$ MQW, (b) a GaN epilayer, (c) an $In_xGa_{1-x}N$/GaN MQW, and (d) an $In_xGa_{1-x}N$ epilayer measured at $T=10$ K.

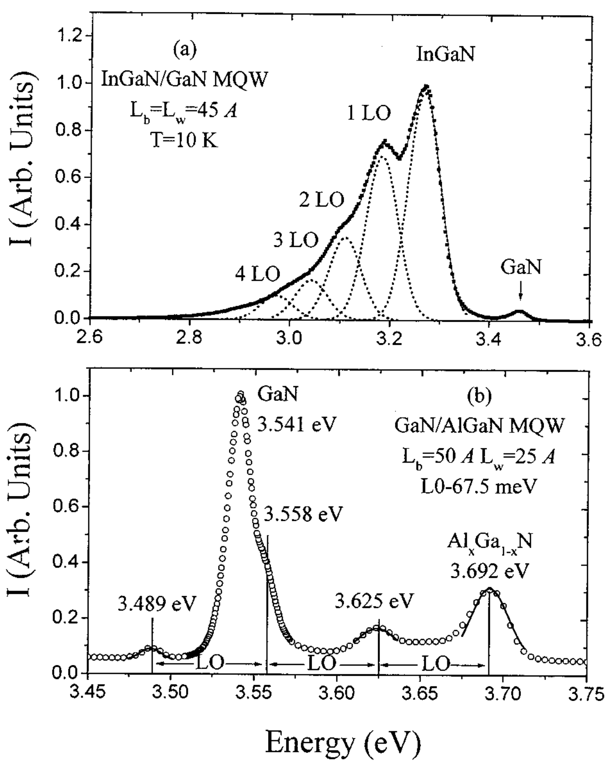

FIG. 2. (a) cw PL spectrum of the 45 Å well $In_xGa_{1-x}N$/GaN MQW measured at $T=10$ K. The solid line is a least squares fit using Gaussian functions (dotted lines). (b) cw PL spectrum of the 25 Å well GaN/$Al_xGa_{1-x}N$ MQW measured at $T=10$ K. Individual peaks were fit using Gaussian functions (solid lines).

pared to the optical transition in the GaN epilayer. The dominating transition line at $T=10$ K at 3.485 eV in GaN epilayer is due to the recombination of the ground state of the A exciton.[8,9] The dominant transition line (at 3.268 eV) for the $In_xGa_{1-x}N$/GaN MQW is due to the localized exciton transition from the $In_xGa_{1-x}N$ well region. Also apparent in the spectrum is an additional peak at 3.481 eV, at the energy corresponding to a transition from the (GaN) barrier region. The main peak from the $In_xGa_{1-x}N$/GaN MQW is blue shifted by about 75 meV compared to that from the $In_xGa_{1-x}N$ epilayer of Fig. 1(d). The dominant transition line for the $In_xGa_{1-x}N$ epilayer, at 3.193 eV, is from the recombination of localized excitons due to alloy disorder.[2]

However, more interesting are the additional features which are clear in the MQW structures, but which are not apparent in either the GaN epilayer, Fig. 1(b), or the $In_xGa_{1-x}N$ epilayer, Fig. 1(d). For the 45 Å well $In_xGa_{1-x}N$/GaN MQW, in addition to the main emission line at 3.268 eV, Fig. 1(c) shows that there are also more features on the low energy side. To illustrate this more clearly, the $T=10$ K cw PL spectrum of the $In_xGa_{1-x}N$/GaN MQW has been replotted in Fig. 2(a). In this plot, we concentrate on the behavior of the well transition. Five emission peaks, at approximately 3.268, 3.181, 3.102, 3.019, and 2.931 eV, have been resolved using a multiple Gaussian fitting function,

$$I(E)=\sum_{i=0}^{4}\frac{A_i}{w_i}\sqrt{\frac{4\ln 2}{\pi}}\exp\left(-\frac{4\ln 2(E_i-E_{c,i})^2}{w_i^2}\right), \qquad (3)$$

where A is the peak area, w is the full width at half-maximum (FWHM), and E_c is the peak energy position. The peak energy positions are separated by 84 ± 4 meV which is close to the longitudinal optical (LO) phonon energy of bulk InN (86 meV).[5] The transition lines at around 3.181, 3.102, 3.019, and 2.931 eV thus correspond to the LO phonon replicas (1, 2, 3, and 4LO) of the main emission line at 3.268 eV.

The additional features evident in the cw PL emission spectra of the GaN/$Al_xGa_{1-x}N$ MQW are shown more clearly in Fig. 2(b). There is no phonon replica of the transition from the GaN well region at 3.541 eV. However, four transition peaks at 3.692, 3.625, 3.558, and 3.489 eV are clearly resolved. We assign these to the zero to three phonon replicas of the excitonic transition resulting from the $Al_xGa_{1-x}N$ barriers. The phonon replica emissions observed in the GaN/$Al_xGa_{1-x}N$ MQW shown in Fig. 2(b) are associated with the LO phonons, except that they vibrate at the GaN bulk TO frequency.[3]

The relative intensities of the phonon replica peaks to the zero-phonon emission line are plotted in Fig. 3, for (a) the $In_xGa_{1-x}N$/GaN and (b) the GaN/$Al_xGa_{1-x}N$ MQW sample. The least squares fit of the data with Eq. (2) for the $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQW are shown as solid lines in Figs. 3(a) and 3(b), respectively. The fitted values of the exciton-phonon coupling constant were found to be $S=0.802$ and 0.556 for the $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQW samples, respectively.

It is interesting to note that (1) neither the high quality GaN nor the high quality $In_xGa_{1-x}N$ epilayer have phonon replicas in their emission spectra as indicated in Figs. 1(b) and 1(d); (2) no phonon replica was observed for the transition from the GaN layer in either MQW, whether as the well layer in the GaN/$Al_xGa_{1-x}N$ MQW or the barrier layer in the

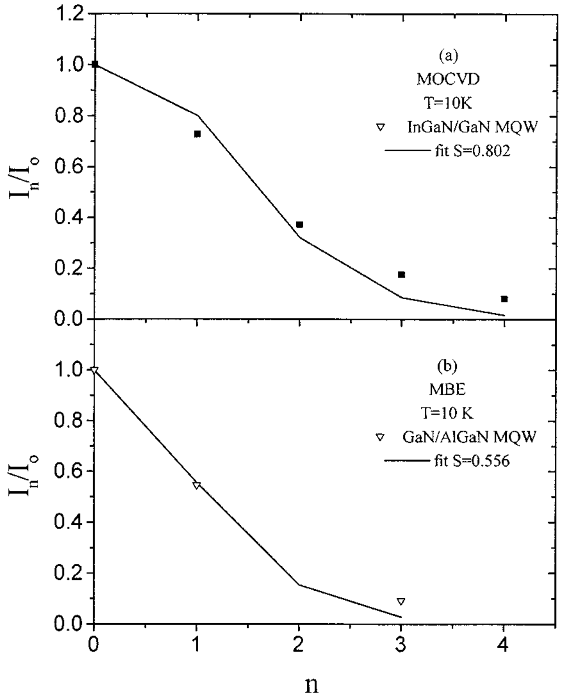

FIG. 3. The ratio of the integrated PL emission intensities of the higher order phonon replica transitions to the zero phonon transition as a function of n, the number of phonons involved, for both (a) the $In_xGa_{1-x}N$/GaN MQW and (b) the GaN/$Al_xGa_{1-x}N$ MQW obtained at 10 K. The relative intensities are fit using Eq. (2) to extract the exciton-phonon coupling constants, which are $S=0.802$ and 0.556 for the $In_xGa_{1-x}N$/GaN and the GaN/$Al_xGa_{1-x}N$ MQW, respectively.

$In_xGa_{1-x}N$/GaN MQW; (3) phonon replicas were only observed for exciton transitions from alloy materials, the InGaN well region in the $In_xGa_{1-x}N$/GaN MQW, and the $Al_xGa_{1-x}N$ barrier region in the GaN/$Al_xGa_{1-x}N$ MQW. Observation (1) indicates that alloy disorder alone (in the $In_xGa_{1-x}N$ epilayer) cannot enhance the exciton-phonon interaction significantly. Similarly, interface roughness alone is also insufficient as indicated by observation (2). The confinement effects on exciton-phonon interaction can also be neglected since phonon replicas were observed from the barrier region in the GaN/$Al_xGa_{1-x}N$ MQW. We thus speculate that the symmetry properties of the MQW together with alloy disorder are responsible for the observation of the phonon replicas in the alloy material regions of the $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQWs. It is well known that phonon properties are modified in MQW compared to that in bulk materials due to the symmetry properties in MQW growth direction.[10] Alloy disorder also seems to be an important contributor to the enhanced exciton-phonon interaction in III-nitride MQW.

It is also interesting to compare our results obtained here to those of the well studied semiconductor CdS which has the same crystal structure (wurtzite) as GaN. A coupling constant S for the exciton-phonon interaction of about 1 to 2 in CdS bulk has been obtained.[11] In contrast, GaN and $In_xGa_{1-x}N$ epilayers which have been studied here show a much smaller coupling constant, since there is little evidence of phonon replicas of the exciton transition in these epilayers. In fact, a smaller coupling constant in GaN epilayers is consistent with a theory developed for CdS materials.[12] It was calculated that the exciton-phonon coupling constant S is inversely proportional to the phonon energy E_p and almost proportional to the ratio of the hole mass to electron mass $a=m_h/m_e$. For CdS, the LO phonon energy E_p is $E_p=37.7$ meV, while the electron and hole effective masses[13] are approximately $m_e^{\parallel}\approx 0.18\ m_0$ and $m_e^{\perp}\approx 0.19\ m_0$, and $m_h^{\perp}\approx 5\ m_0$ and $m_h^{\perp}\approx 0.7\ m_0$, respectively, where $m^{\parallel}$ and $m^{\perp}$ are the effective masses in the directions parallel and perpendicular to the c axis of the crystals. For GaN,[5,14] the LO phonon energy $E_p=91$ meV, $m_e^{\parallel}\approx m_e^{\perp}\approx 0.19\ m_0$, $m_h^{\parallel}\approx 2\ m_0$, and $m_h^{\parallel}\approx 0.33\ m_0$. From these numbers, we expect the exciton-phonon coupling constant S of the bulk GaN to be approximately a factor of six smaller than that of CdS due to its larger LO phonon energy and its smaller effective mass ratio of the hole to the electron. On the other hand, the coupling constant is expected to be changed in MQW structures due to the modified phonon dispersion relation in MQWs.

In summary, the exciton-phonon coupling has been studied in $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQWs. The coupling was found to be enhanced in alloy regions of the quantum wells compared to epilayers due to a combination of the symmetry properties of MQW and the effects of alloy disorder. The coupling constants S were extracted for the $In_xGa_{1-x}N$/GaN and the GaN/$Al_xGa_{1-x}N$ MQWs, and were found to be $S=0.802$ and 0.556, respectively. It is well known that the carrier-phonon interaction is one of the important factors that determines the optical transition spectra and the gain mechanism for lasing. The enhanced exciton-phonon interaction in the $In_xGa_{1-x}N$/GaN and GaN/$Al_xGa_{1-x}N$ MQW systems will play an important role for understanding the mechanisms of optical transitions in many GaN based optoelectronic devices, including carrier relaxation, transformation, and recombination dynamics as well as LED emission and laser mechanisms.

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