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Thicknes dependence of temperature-induced emission mechanism in InGaN/AlGaN short-period superlattices

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An analysis of temperature-dependent photoluminescence (PL) spectra for a series of InGaN/AlGaN short-period superlattices (SP-SLs) with different well and barrier thickness is presented. A quantitative model, based on Gaussian-like function of localized electronic states, to fit the temperature-dependent emission peak energy gives good fits over an extended temperature range for all samples. It is found that, among all parameters in the model, the degree of broadening of the Gaussian distribution is strongly dependent on the structural parameters of SP-SLs and determines the anomalous “S-shape” behavior of the temperature-dependent emission energy. In thin well and barrier samples with higher broadening parameter, the temperature-dependence of emission energy is different from those of typical “S-shape” behavior, which is characterized by the bigger red-shift with no blue shift in the temperature range used. The depth of localization, $E_a-E_0$, is smaller than the corresponding activation energy obtained from the thermal quenching of the PL intensity, thus, indicating that the thermal quenching activation energy and the localization due to band-gap fluctuation most likely have different origins. We demonstrate that, in the InGaN/AlGaN SP-SLs, the interface characteristics also contributes to the temperature-induced PL emission shift as much as the compositional fluctuation does. © 2012 American Institute of Physics.

I. INTRODUCTION

Short-period superlattices (SP-SLs) serve as important elements of a hetero-structural design, aimed at solving either technological or design problems.1–3 The specific optical and electronic properties of SP-SLs are determined by the existence of mini-gaps in the conduction and valence bands. SP-SLs can be considered, therefore, as a bulk material with anisotropic properties which are controlled by its internal microscopic parameters—thicknesses and compositions of constituent layers. Among them, the SP-SLs composed of InGaN and AlGaN layers can be used to replace quaternary In$_x$Al$_{1-x}$Ga$_y$N layers in active regions in light emitting diodes and laser diodes, in order to avoid the detrimental effects often observed in bulk In$_x$Al$_{1-x}$Ga$_y$N layers while maintaining their advantages of independently adjusting the band gap and lattice constant by varying the composition and thickness of consisting layers.4,5

One of the most characteristic features in InGaN alloys and quantum wells is the so-called “S-shape,” used to describe the anomalous behavior of the photoluminescence (PL) emission peak as a function of temperature. There are several different explanations for this decrease-increase-decrease pattern which is distinguishable from the usual behavior of semiconductor materials, typically known as Varshini’s.6 Narukawa et al.7 have suggested that recombination in InGaN wells could occur in In-rich regions acting as quantum dots. Other authors have attributed the emission to the recombination of excitons localized either by In compositional fluctuations or at the band tail states.8,9 It has also been proposed that the cause of the temperature-induced “S-shape” PL shift is a change in the carrier recombination dynamics due to inhomogeneous energy states, and a quantitative model has been given assuming that the density of states distribution is described by a Gaussian distribution.10 More comprehensive model11 is proposed by combining the empirical relation for the temperature-dependence of the fundamental energy band gap into the existing model given in Ref. 10.

In this paper, temperature-dependent photoluminescence spectra of InGaN/AlGaN SP-SLs have been measured, especially the observed anomalous behavior of the luminescence peak positions and intensities of the samples with different layer thicknesses. A model considering thermally activated redistribution within a Gaussian distribution of localized states and the temperature dependence of fundamental band gap is used quantitatively to describe the emission behavior of the samples over the entire temperature range in terms of localization energies, inhomogeneous broadening, and carrier lifetime in SP-SLs. The dependence of these parameters on the change of layer thickness allows us to understand the emission behavior of InGaN/AlGaN SP-SLs and to give an idea in real applications.

II. EXPERIMENTAL PROCEDURES

The epitaxial structures of InGaN/AlGaN SP-SLs used in this study were grown on c-plane (0001) sapphire substrates.
by metal organic chemical vapor deposition (MOCVD) equipment, following the growth of a 4 μm thick undoped buffer layer. Two sets of In$_{0.13}$Ga$_{0.87}$N/Al$_{0.065}$Ga$_{0.935}$N SP-SL layers with different layer thickness were grown. In the first set of samples (A1–A3), the thickness of In$_{0.13}$Ga$_{0.87}$N layer varies from 7.8 Å to 23.2 Å while that of Al$_{0.065}$Ga$_{0.935}$N barrier layer is kept constant to 15 Å. The second set of samples (B1–B4) is prepared with different Al$_{0.065}$Ga$_{0.935}$N thickness ranging from 11 Å to 54 Å while keeping the same thickness of 11.3 Å In$_{0.13}$Ga$_{0.87}$N layer. The total thickness of all SP-SL samples was maintained constant 3000 Å. Structural parameters of samples including the composition, lattice tilt, strain condition, and quality have been obtained by high-resolution ω-2θ x-ray diffraction and reciprocal space mapping (RSM) measurements. In this study, RSM is prepared based on the asymmetric x-ray diffraction in the (1015) direction. The fitting of the experimental XRD/RSM results is performed with the simulation based on LEPTOS software, Bruker-AXS. The extracted layer thicknesses of each sample were confirmed by high-resolution transmission electron microscopy (HR-TEM). HR-TEM images were obtained using a JEOL 4000EX operating at 400 kV.

Temperature-dependent PL spectra were measured using a continuous wave (cw) He-Cd (20 mW) laser with a wavelength of 325 nm as the exciting source. The samples were mounted in a closed-cycle helium cryostat where the temperature was varied from 20 K to 300 K. The luminescence was dispersed by a monochrometer and detected by a photomultiplier using a standard lock-in technique.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the HR-XRD ω-2θ scans of the first set In$_{0.13}$Ga$_{0.87}$N/Al$_{0.065}$Ga$_{0.935}$N SP-SL samples with different InGaN thickness. The zero-order and higher-order satellite diffraction peaks are clearly distinguishable in all samples, indicating that the good integrity and periodicity of superlattice are well maintained. Small-amplitude interferences, or Pendellosung fringes, are seen on either side of the substrate peak, in which the angular separation between two interference fringes maxima is related to the total epi-layer thickness. The strain of the superlattice structure is determined by measuring a RSM of diffraction from an asymmetric plane, as shown in Figure 1(b). The reciprocal space maps with asymmetric plane of $(hkl)$ is illustrated with contour distribution in the rectangular coordinate $q_x = 2\pi \sqrt{\frac{4}{a^2} (h^2 + hk + k^2)}$ and $q_z = \frac{\lambda}{2l}$, where $a$ and $c$ are the lattice spacing in the a-plane and c-plane, respectively. For all samples of both set, the observed diffraction maxima of each sample share the same value for the parallel component $q_x$, then all layers must have the same in-plane lattice constant and the structure is fully strained.

The 0-th order peak position and the spacing of higher-order peaks are used to calculate the average composition and the period of each superlattice. The average lattice constant $\bar{c}$ plays an important role in the characterization, since it equals the average of the lattice constants weighted with the related thicknesses. In other words, knowledge of the values of $\bar{c}$ and thickness yields the remaining unknown lattice constant. The thicknesses of the individual layers and the out-of-plane lattice constants are linked together by the following equation:

$$\bar{c} = \frac{\lambda}{\sin(\theta_{SLO})} = \frac{c_1 t_1 + c_2 t_2}{t_1 + t_2},$$

Figure 1. High-resolution ω-2θ diffraction profile of the first set of SP-SL samples (A1, A2, and A3) near (0004) GaN peak. (b) RSM contour of In$_{0.13}$Ga$_{0.87}$N (11.3 Å)/Al$_{0.065}$Ga$_{0.935}$N (15 Å) SP-SLs (sample A2). The almost vertical dashed line, which connects the centers of the successive elliptical areas, implies that the sample is fully strained. (c) High-resolution TEM pictures of sample A2 for the comparison purpose.
where \( \theta_{SLO} \) is the angular position of the zero-order satellite peak, \( c \) and \( t \) are the lattice constants and thicknesses for materials 1 and 2. The above equation states that the average lattice constant is directly determined from the symmetric XRD scan. The total thickness of the repeated SL unit, \( t_1 + t_2 \), can be calculated by using the angular separation between the first-order peaks, which are arrayed symmetrically with respect to zero-order peak on both side of the rocking curve using the equation

\[
\text{Period} = \left( \frac{n_i - n_j}{2} \right) \frac{\lambda}{\sin \theta_i - \sin \theta_j},
\]

where \( n_i \) and \( n_j \) show the satellite peak order and \( \theta_i \) and \( \theta_j \) are the diffraction angles. The compositions of ternary compounds are determined by utilizing Vegard’s law and the previously established material parameters.\textsuperscript{12} HR-TEM measurements are also used to verify the thicknesses obtaining in HR-XRD measurements, as shown in Figure 1(c). The extracted composition and thickness of the constituents of the samples are summarized in Table I.

Figure 2 shows PL spectra at different temperatures for sample A3 with 22.3 Å In\(_{0.13}\)Ga\(_{0.87}\)N whose emission peak energy is \( \sim 3.13 \) eV at 20 K, and the inset shows an Arrenhius plot of the normalized PL intensity for the SP-SL related PL emission over the temperature range under investigation. As the temperature increases from 20 to 10 K, the peak energy redshifts 16 meV, which is much larger than the expected band-gap shrinkage of 4 meV over this temperature range. In contrast, the PL peak blueshifts 10 meV between 100 and 160 K. By considering the estimated temperature-induced bandgap shrinkage of 13 meV, the actual blueshift of the PL peak with respect to the band edge is about 23 meV over this temperature range. After that, the emission energy is redshifted again with increasing temperature. In this high temperature range, a non-radiative thermal quenching of the PL intensity with an activation energy of 60.6 meV is observed.

Figures 3 and 4 show the position of the main PL peak as a function of temperature for the first and second sets of samples, respectively. The emission energy of the sample A1 with thin 7.8 Å In\(_{0.13}\)Ga\(_{0.87}\)N well is redshifted with increasing temperature up to 300 K. The emission energy of the sample A2, in which the thickness of In\(_{0.13}\)Ga\(_{0.87}\)N is 11.32 Å, is redshifted up to around 200 K, and is then blueshifted with further increase in temperature up to about 300 K. On the other hand, the blueshift of emission energy of the sample A3 with 23.2 Å thick In\(_{0.13}\)Ga\(_{0.87}\)N well, occurs at lower temperature near 100 K, and then, the emission energy is redshifted again with the increase of temperature. This temperature-induced “S-shaped” PL shift is typical in InGaN-related emission with increasing temperature, and is attributed to the localized exciton emission. The transition temperatures from redshift to blueshift move towards lower temperatures as the thickness of InGaN increases. The same trend is also observed in the second set of samples where the transition from redshift to blue-shift occurs at lower temperatures as the thickness of AlGaN barrier increases. The observed result implies that the localization of radiative electron-hole recombination is strongly dependent on the structure of SP-SLs.

The “S-shape” behavior is conventionally interpreted as a sign of localization due to inhomogeneities of the potential and the carrier localization in its band tail states, due to the fluctuations in In composition, layer thickness and possibly defects in the heterostructure. It is thus of interest to consider how the behavior and these effects vary with the structural parameters, i.e., the constituent layer thickness in this study. Recently, a quantitative model considering a Gaussian bandtail distribution of localized states and the temperature dependence of the fundamental bandgap is given as following:\textsuperscript{11}:

\[
E_{PL} = E_{PL(0)} - S(E_{ph}) \left[ \coth \left( \frac{E_{ph}}{2k_BT} \right) - 1 \right] - x(T)k_BT, \tag{3}
\]

where \( k_B \) is the Boltzmann’s constant, \( E_{PL(0)} \) is the emission peak energy at 0 K, \( S \) is related to the Huang-Rhys parameters,\textsuperscript{13} a dimensionless coupling constant that represents the strength with which the radiative transition couples with the LO polarization field, \( \langle E_{ph} \rangle \) is the average phonon energy of GaN (\( \sim 9 \) meV)\textsuperscript{14} and \( x(T) \) is a dimensionless coefficient that can be obtained by numerically solving the equation

\[
x e^x = \gamma_1 \left[ \left( \frac{\sigma}{k_BT} \right)^2 - x \right] e^{-(E_a - E_v)/k_BT}, \tag{4}
\]

where \( E_a - E_v \) is the barrier height that carriers must overcome to transfer and represents an averaged value of the depth of localization, \( \gamma_1 \) represents the ratio of the carrier recombination time to the carrier transfer time (\( \tau_i/\tau_{tr} \)), and

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TABLE I. Structural parameters of (In\(_{x}\)Ga\(_{1-x}\)N)\(_{n}\)/(Al\(_{y}\)Ga\(_{1-y}\)N)\(_{m}\) superlattices obtained from the high-resolution XRD and HR-TEM measurements. The model parameters extracted from the fitting of the temperature-dependent emission peak energies to Eqs. (3) and (4) are summarized in the right column.
\( \sigma \) is the width of the Gaussian distribution of localized states, 
\[ \rho(E) \propto \exp\left[-\frac{(E - E_o)^2}{2\sigma^2}\right], \]
where \( E_o \) and \( \sigma \) represent the center and width of the distribution function, respectively.

Equations (3) and (4) are employed to numerically fit the experimental data of the temperature-dependent emission peak energies for the first and second set of samples, shown as solid lines in Figs. 3 and 4, respectively. The good agreement between theory and experimental data over the entire temperature range is obtained. The fitting parameters are summarized and are listed in Table I.

The depth of localization, \( E_a - E_o \), reflects the magnitude of the carrier localization, which is in the range between 40 meV and 50 meV in most samples. It is known that the increase in indium content increases this depth. In the case of InGaN MQWs, for instance, values of 60 meV have been reported for samples emitting at 2.9 eV.\(^{15}\) Therefore, the observed depth of 40-50 meV in the samples emitting near 3.1 eV is in good agreement with the trends reported here. On the other hand, when well thickness is down to 7.8 Å, the smaller energy difference of 29.9 meV is observed even though the same indium content is used in the well. It indicates that, when the same well indium composition is used, the compositional fluctuation is less severe in thinner wells and it is somehow related to the amount of well strain.

As shown in the inset of Fig. 2 for sample A3, the temperature dependence of the integrated PL intensity for all samples is examined, and the activation energies of \( E_A \) for all samples are obtained by fitting the data with \( I \propto \exp\left(-E_A/k_BT\right) \) and are listed in Table I. In general, the quenching of the luminescence with temperature can be explained by thermal emission of the carriers out of a confining potential with an activation energy correlated with the depth of the confining potential. Since the observed activation energy is much less than the band offsets as well as the band gap difference between the wells and the barriers, the thermal quenching of the InGaN-related emission is not due to the thermal activation of electrons and holes from the InGaN wells into the AlGaN barriers. Instead, the dominant mechanism leading to the quenching of the InGaN-related PL is due to the thermionic emission of the photocarriers out of the potential minima caused by potential fluctuations. Our analysis clearly shows that the localization energy and band-gap fluctuation \( E_a - E_o \) in all of our samples are smaller than \( E_A \). Furthermore, the PL line shape indicates that carriers are free and thermalized for \( T \geq 120 \text{ K} \) which is higher than the temperature range in which the blueshift due to the depth of localization occurs. Thus, the thermal quenching
activation energy must represent either a barrier to capture at non-radiative recombination centers, or the thermal activation energy of such centers. It is interesting to note that the increase in $E_A$ is observed in the samples with thinner InGaN wells in the first set and thinner AlGaN barriers in the second set.

The broadening parameter $\sigma$, the width of the Gaussian distribution of localized states representing the broadening of the absorption edge, becomes greater as the well thickness and the barrier thickness decrease. It has been known that the absorption edge tends to increase as the indium content increases. Since the same well composition is used in all samples, it seems that the influence of another factor must be considered. It is speculated that interface potential fluctuations at the rough interface may cause the inhomogeneous broadening of the absorption edge since the wave function penetration across the interface is more severe in thinner well and barrier quantum wells. Therefore, SP-SLs with thin wells and barriers such as samples A1 and B1, where the broadening parameters are bigger than the rest of samples, redshift faster than that predicted by Varshni’s empirical formula as the temperature increases and show different shape in the temperature dependence of emission energies from the typical “S-shape” curve. In the case of SP-SLs, the increase in the broadening parameter $\sigma$ with the activation energy $E_A$ in thermal quenching represents that both parameters have the same origin due to inhomogeneous interface containing non-radiative centers.

Considering the carrier transfer time as approximately constant in samples with same barrier thickness (A1–A3), the recombination time is a strong variation with different well thicknesses. Recombination time is enhanced in thicker quantum wells where the piezoelectric field is bigger due to the enhancement of quantum confinement Stark effect (QCSE). The same trend of enhanced recombination time in thicker well is also reported in other literature. On the other hand, the ratio $\gamma$, is seen to decrease with the barrier thickness increases. This is considered due to the longer transit time in thicker barrier SP-SLs.

Thus, the action of intense piezoelectric fields across the well and the barrier coupled with the inhomogeneous well material has an important effect on the parameters fitted by the model given by Eqs. (3) and (4).

IV. CONCLUSIONS

In conclusion, we have investigated the so-called “S-shape” behavior in a number of InGaN/AlGaN SP-SLs with different well and barrier thicknesses. We have fitted the temperature-dependent PL emission peak energy of a series of samples to a model that takes into account fundamental parameters that characterize the “S-shape” and describe the variation of the localization depth, broadening with emission energy and the ratio of the carrier recombination time to transfer time between localized levels. The emission mechanism of InGaN/AlGaN SP-SLs is strongly related to the fluctuations at the interface and in the InN fraction, and its distribution in addition to the strain applied to the superlattice structure.

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