Effect of multilayer barriers on the optical properties of GaInNAs single quantum-well structures grown by metalorganic vapor phase epitaxy

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We report on the effects of combined strain-compensating and strain-mediating layers of various widths on the optical properties of 1.3 μm GaInNAs/GaAs single quantum well structures grown by metalorganic vapor phase epitaxy (MOVPE). While the emission wavelength of GaInNAs/GaAs quantum wells can be redshifted by the adoption of strain-compensated GaNAs layers, the material quality is degraded by the increased stress at the well/barrier interface. This detrimental effect can be cured by inserting a strain-mediating InGaAs layer between them. Contrary to what is expected, however, the emission wavelength is blueshifted by the insertion of the InGaAs layer, which is attributed to the reduced N incorporation due to the improved interface quality. Our results indicate that the optical properties of MOVPE-grown GaInNAs/GaAs quantum wells can be optimized in quantum efficiency and emission wavelength by combination of strain-compensating and strain-mediating layers with suitable characteristics.
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Dilute-nitride III–V compound semiconductors and their related heterostructures have been recognized as very promising materials for optoelectronic devices, especially in the 1.3–1.6 μm wavelength range.[1–6] Various devices with performance based on GaInNAs/GaAs heterostructures have been developed.[2–6] Efficient light-emission is crucial for many such device applications. However, incorporation of N into InGaAs decreases the radiative transition matrix elements, which will increase the threshold in the case of lasing,[7] and the luminescence efficiency in GaInNAs also decreases rapidly with increase of N content due to the increase in nonradiative recombination centers. The solubility of N in these materials is quite limited due to the large miscibility gap between As and N, which in turn leads to difficulty in incorporating N, especially in metalorganic vapor phase epitaxy (MOVPE). It turns out that low N content is preferred when growing GaInNAs materials, although the introduction of N should be favorable for the relief of macroscopic compressive strain in InGaAs with high In content. In order to assure long emission wavelength as well as high emission efficiency, adoption of strain-compensating GaNAs layers (SCLs) and strain-mediating GaInNAs layers (SMLs) has proved an effective approach in molecular beam epitaxy (MBE),[7] but further studies are needed on how such layers influence the growth and optical properties of the heterostructures. In this letter, we investigate the effect of GaNAs SCL and GaInAs SML with varying widths on the optical properties of MOVPE-grown GaInNAs/GaAs single quantum well structures by using photoluminescence (PL) and PL excitation (PLE) spectroscopy.

The samples used in this study are grown by MOVPE in a horizontal-type reactor at low pressure (100 mbar) on epiready (001) GaAs substrates. The basic SQW structure is an 8 nm $Ga_{0.7}In_{0.3}N_yAs_{1-y}$ QW between GaAs barriers. $GaN_{0.2}As$ SCLs and $Ga_{0.7}In_{0.3}As$ SMLs with various thicknesses have been inserted between the $Ga_{0.7}In_{0.3}NAs$ QW and GaAs barriers as shown in the inset of Fig. 1. Triethylgallium (TEG), trimethylindium (TMI), tertiarybutylarsine (TBA), and 1,1-dimethyl hydrazine (DMHy) were used as respective precursors for the sources of Ga, In, As, and N, to grow the GaInNAs layers. The flow ratio of group V to group III is about 5. A 100 nm GaAs capping layer was grown at the QW growth temperature of 470 °C. All GaInNAs QW samples were *in situ* annealed at 650 °C for 10 min in AsH_3 ambient. The growth conditions are identical for all samples in this study except for the thickness of GaNAs and InGaAs layers. For PL measurements, the samples were excited by a high power diode laser (670 nm). The excitation light source for PLE measurements was a 250 W tungsten-halogen lamp combined with a 0.27-m-grating monochromator and suitable filters.[8]

Figure 1 compares the PL spectra taken at room temperature of four samples with different thicknesses of SCL

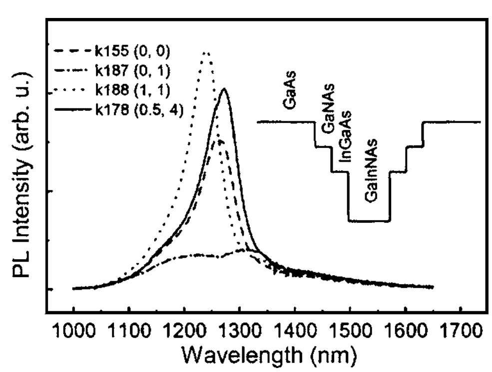

FIG. 1. PL spectra taken at room temperature for QW samples with different thickness SMLs and SCLs.

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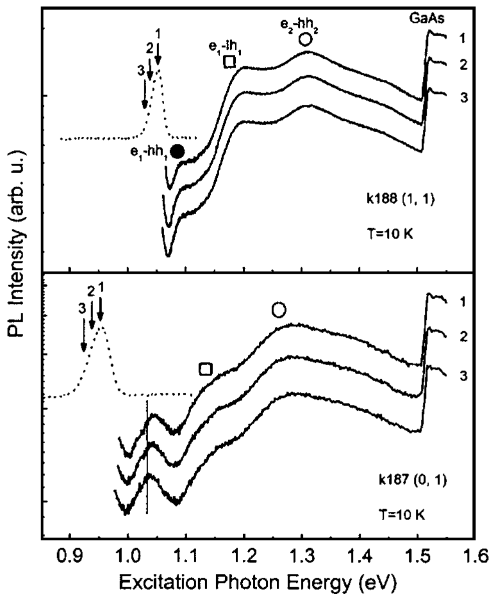

FIG. 2. Spectra of PL and PLE at 10 K for samples k 187(0,1) and 188 (1,1).

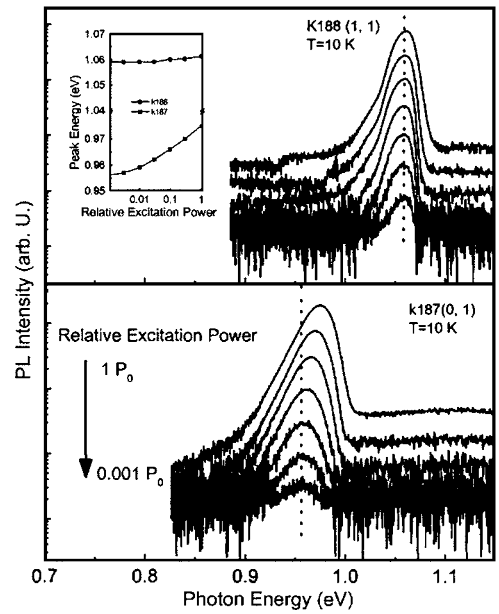

FIG. 3. Dependence of PL spectra at 10 K on excitation power for samples k 187(0,1) and 188 (1,1). The highest power P_0 is ~84 mW. The dashed straight lines are a guide for the eye. The inset is the energy of the PL peak position as a function of relative excitation power for the two samples.

and SML. For simplicity, the corresponding thickness of InGaAs SML and GaNAs SCL is denoted as (SML, SCL) in units of nanometers. The peak wavelength of reference sample k155 (0,0) is 1263 nm. With the introduction of GaNAs SCL [sample k187 (0,1)], the PL peak wavelength shifts to 1317 nm. It is also noted that, as well as the redshift, the PL intensity has decreased significantly and the PL linewidth is greatly broadened, and there appears a strong PL component on the short wavelength side. After further insertion of 1 nm InGaAs SMLs between the GaNAs SCL and GaInNAs QW, the optical properties of sample k188 (1,1) is apparently improved, represented by the substantial increase of PL intensity and the narrowing of the PL linewidth. Therefore it is concluded that the degradation in optical properties by GaNAs SCLs can be effectively surmounted by a thin InGaAs SML. However, it is interesting to note that the PL emission in k188 is shifted to even shorter than the sample without SCL and SML. This is contrary to what has been observed in MBE-grown samples,[7] the reason for which will be clarified below. Nevertheless, another sample k178 (0.5,4) has demonstrated increased PL intensity as well as longer wavelength compared to sample k155 (0,0). These results indicate that optical properties of a GaInNAs QW can be adjusted by employing the combined SML and SCL with suitably adjusted thicknesses. The overall improvements in material quality possible by this approach have been demonstrated by our recent demonstration of a MOVPE-grown 1360 nm laser diode with threshold current density of 892 A/cm^2.[9]

In order to better understand the effects of SMLs and SCLs on the optical properties of GaInNAs QWs, we have performed detailed PL and PLE measurements. Figure 2 shows the PL and PLE spectra for two samples, where PLE spectra with different detection energy have been plotted. As can be seen, sample k188 (1,1) exhibited absorption features typical for a QW, namely, step-like density of states and independence of spectra on detection energy. The transitions denoted in the figure can be attributed to e_1–hh_1, e_1–lh_1, and e_2–hh_2, respectively. In comparison, sample k187 (0,1) demonstrates similar feature in the higher energy range although the corresponding transition energies are lower. However, near the transition edge of e_1–hh_1 transition, the PLE spectrum exhibits a "resonant-like" shape and the resonance peak position shifts with the detection energy. This result indicates that the sample with only an SCL should not be considered as a normal QW, but rather as a system with some deeply localized states embedded in the QW plane.[10] In this case, the PL at low temperature originates from the ground localized states while the resonant feature in PLE corresponds to excited localized states which are just below the extended e_1–hh_1 QW transition and overlap with the QW absorption edge. When the excitation energy is greater than the lowest extended QW transition, the excited carriers are mobile in the whole QW plane and randomly captured by the localized states, which can explain the detection-energy-independence of PLE at higher energy. When the excitation energy is lower than the lowest extended QW state, the photons only excite carriers (excitons at low temperatures) resonantly in particular "islands" with appropriate excited levels. These carriers at excited localized states then relax inside the "islands" to the ground localized states where they recombine radiatively. The dependence of PLE at lower energy on the detection energy results from the energy selectivity of particular "islands." This strong localization effect has increased the inhomogeneous broadening represented by the broader PL width and bigger Stokes shift, as demonstrated in Fig. 1. Comparatively, much weaker localization effects occur in k188(1,1).

The formation of deeply localized states in k187(0,1) is further evidenced by the dependence of PL on the excitation intensity. Shown in Fig. 3 is the comparison of excitation intensity dependence of PL for samples k187 (0,1) and k188 (1,1). For the sample with only GaNAs SCLs, the PL maximum is sensitively dependent on the excitation intensity. The PL maximum shifts ~19 meV towards the higher energy when the excitation intensity increases over three orders. In comparison, the sample with further insertion of an InGaAs SML shifts only 2 meV. The phenomenon that PL shifts in energy with excitation intensity can be interpreted by band filling of localized states.[8] The magnitude of the shifts should be related to the distribution of density of states; the shift will be larger if the emission is due to deeply localized states because they are far away from the QW transition edge.

It is apparent from the difference in the optical characterizations of samples k187(0,1) and k188 (1,1) that the optical quality of GaInNAs QWs is directly related to the characteristics of the well/barrier interface. For the GaNAs/GaInNAs interface, the enlarged difference in lattice constant may result in structural defects at the interface or partial phase separation in the QW,[11,12] which can account for the strong localization. The observed emission band in the short wavelength side in k187 (0,1) may relate to defect-related states generated in the QW/barrier interfaces.[11] In addition, the structural quality of the GaNAs may be deteriorated by the existence of N which also influences the subsequent growth of the GaInNAs QW. Both of the above effects are inherent properties of dilute nitrides[12] and could be cured by InGaAs SMLs.

Returning to the PL wavelength, it is expected that the band gap of QWs of fixed composition should decrease after the insertion of GaNAs and or InGaAs layers due to the effective broadening in well width. We have pointed out that although the PL wavelength in our studies redshifted due to the introduction of GaNAs SCLs, it blueshifted with further introduction of InGaAs SMLs. We attribute this "anomaly" to the difference in N contents in the two samples. Although it is hard to determine the QW ground state energies exactly due to the overlap with the excited localized states in Fig. 2, the increase of other transition energies (e_1–lh_1 and e_2–hh_2) implies that the N content in sample k188 (1,1) should be higher than in k187 (0,1). Taking into account the above discussion of the structural quality related to the SMLs and SCLs, it is suggested that incorporation rate of N in GaInNAs QWs in MOVPE is correlated to the quality of the interfaces: the higher the interface quality the lower the N content. Actually, we found that the PL wavelength is blueshifted on increasing the thickness of SMLs between 0.5 and 2 nm if fixing the SCLs at 1 nm. This can be interpreted by the improvement of the poor interface between GaNAs and GaInNAs with increasing the thickness of InGaAs. The fact that the incorporation rate of N is dependent on the quality of the interface may account for the varied observations on this issue in MOVPE, but the mechanisms need further study.

Finally it is interesting to examine the PL wavelength as a function of the thickness of GaNAs SCLs with fixed 0.5 nm InGaAs SML, as shown in Fig. 4. Again contrary to what is expected, the PL wavelength decreases with increase in the thickness of the SCLs. This result may imply that more-compensated strain is favorable for the reduction of structural defects in compressively strained InGaAs and GaInNAs layers and therefore decreases the incorporation of N. One may argue that the change of strain may affect the transition energy. However, if it does, the more tensile strain in the thicker GaNAs layers should lower the energy of e_1–hh_1 transition in the QW.

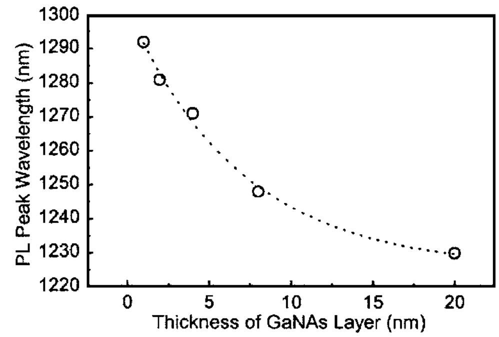

FIG. 4. PL peak wavelength as a function of the thickness of GaNAs SCLs for a series of samples with 0.5 nm InGaAs SMLs. The broken curve is a guide for the eye.

In conclusion, we have investigated the dependence of the optical properties of GaInNAs QWs grown by MOVPE containing GaNAs SCLs and InGaAs SMLs. Introduction of only SCLs between the GaAs barriers and GaInNAs QW deteriorates the epitaxial quality of the QW due to the increased stress between tensile-strained GaNAs and compressive-strained GaInNAs. This detrimental effect of the interface can be obviated by the insertion of a layer of InGaAs. Contrary to what observed in MBE, MOVPE-grown GaInNAs QW with combined SCLs/SMLs demonstrates blueshifted PL due to the decreased N content with improved QW quality.

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