Metalorganic molecular-beam epitaxy and characterization of GaAsNSe/GaAs superlattices emitting around 1.5-μm-wavelength region

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GaAsNSe/GaAs superlattices (SLs) were grown on GaAs(001) substrates by metalorganic molecular-beam epitaxy. Strong photoluminesence (PL) emission around the 1.5-μm-wavelength region was observed from these SLs without thermal annealing, which suggests that high electron concentrations in GaAsNSe layers increase the radiative recombination rate, making the nonradiative recombination relatively unimportant. It is also demonstrated that (GaAsNSe/GaAs SLs)/(GaAsN/GaAsSb SLs)/GaAs heterostructures are effective to reduce the strain accumulation in the layers, which will also form effective separated confinement heterostructure. The temperature dependence of the PL peak intensity is drastically improved by combining the GaAsN/GaAsSb SLs with the GaAsNSe/GaAs SLs following this scheme. The PL peak intensity observed at 300 K was as large as 20% of that observed at 19 K. This improvement of the optical property will be attributed to the elimination of nonradiative defects by minimizing average strain in the samples. [DOI: 10.1063/1.1544656]

To realize future large-scale optical-fiber networks, it is crucial to have wavelength division multiplexing (WDM) systems for increasing the number of channels. Spectrum-slicing techniques, in which a broad spectrum is sliced by narrow-band optical filter, have been intensively investigated to realize WDM systems.[1] Superluminescent diodes (SLDs) are desirable for light sources in the spectrum-slicing technique. SLDs 1.3 and 1.55 μm in wavelength have been developed using GaInAsP/InP systems on InP substrates. A tapered GaInAsP/InP SLD emitting in a 1.55-μm-wavelength region was reported recently.[2] A chirped quantum-well structure with different well thickness in the active layer was introduced for increasing the spectral width to as large as 60 nm. However, in conventional GaInAsP/InP systems, there remains the temperature stability issue due to the poor electron confinement resulting from the small conduction band offset.[3,4]

In this regard, GaInAsN/GaAs has received considerable attention as a material system for long-wavelength devices, because it has a large conduction band offset.[5] GaInAsN edge-emitting lasers with higher characteristic temperature and vertical-cavity surface-emitting lasers operating around 1.3 μm have been demonstrated.[6,7] To achieve the more longer wavelength emission, high-quality GaInAsN active layers with larger-N composition have to be grown. However, it still remains difficult due to a large miscibility gap problem to grow the ternary alloys.

In an earlier report, quaternary GaAsNSe alloys grown on GaAs substrates by metalorganic molecular-beam epitaxy (MOMBE) were reported.[8] It was found that the presence of Se during the growth significantly increased the N incorporation in the grown layers, which resulted in a dramatic reduction of the band-gap energy. Since the GaAsNSe alloys were heavily doped with donors up to $\sim 1\times 10^{20}$ cm^{-3}, degenerate Fermi statistics will be applied in the conduction band and nonalloyed ohmic contact was realized with this new alloy semiconductor.[8]

In this letter, optical properties of the GaAsNSe alloys are examined. The samples prepared are in the form of GaAsNSe/GaAs superlattices (SLs) grown on GaAs(001) substrates. Strong photoluminescence (PL) around a wavelength of 1.5 μm is observed from these SLs without thermal annealing, which is usually inevitable to observe intense PL in these N-containing III–V–N semiconductors. It is also demonstrated that the temperature dependence of the PL intensity is drastically improved by combining GaAsN/GaAsSb SLs with the GaAsNSe/GaAs SLs on both sides. The mechanism will be discussed from the viewpoint of residual average strain in the heterostructure.

The samples were grown on GaAs(001) substrates by MOMBE. The precursors used were triethylgallium, trisdimethylaminoarsenic (TDMAAs), monomethylhydrazine, trisdimethylaminoantimon, and ditertiarybutylselenide. The growth temperature was 540–600 °C. The GaAs surfaces were cleaned by heating them to 600 °C for 15 min with the simultaneous supply of TDMAAs, and GaAs buffer layers were grown on the substrates. GaAsNSe/GaAs SLs or (GaAsN/GaAsSb SL)/(GaAsNSe/GaAs SL)/(GaAsN/GaAsSb SL) structures with different SL periods were then grown. Detailed growth conditions of the GaAsNSe layers and their electrical properties were reported elsewhere.[8] SL periods and alloy compositions for the samples are summarized in Table I. The layers of the GaAsNSe/GaAs SL in the samples A and B consist of a 16-nm-thick GaAsNSe layer

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TABLE I. Sample structures: M_1 and M_2 are periods of GaAsNSe/GaAs SLs and GaAsN/GaAsSb SLs, respectively.

	GaAsNSe/GaAs SL			GaAsN/GaAsSb SL			
Sample	M_1 periods	N (%)	Se (%)	M_2 periods	N (%)	Sb (%)	Average strain ($\times 10^{-3}$)
A	10	2.2	2.9	0	0	0	−8.9
B	5	1.2	7.6	0	0	0	−8.7
C	11	2.2	2.9	17	0.5	11	+0.9
D	5	2.2	2.9	5	0.5	11	−0.8

and a 3.4-nm-thick GaAs layer. In the samples C and D, GaAsNSe/GaAs SLs were sandwiched by GaAsN/GaAsSb SL layers. The thickness of both GaAsN and GaAsSb layers is 3.2 nm. The lattice constants and SL structures of the samples were examined by high-resolution x-ray diffraction (XRD) measurements. The PL characteristics of the samples were measured in the temperature range of 19–300 K. All the samples studied in this letter are as-grown and are not treated with conventional thermal annealing.

Figure 1 shows a typical XRD profile of the ten periods of GaAsNSe/GaAs SL grown on a GaAs substrate (sample A). Sharp, intense satellite peaks and interference fringes diffracted from the ten periods in the structure can be clearly seen, which suggests that high-quality GaAsNSe/GaAs SL was grown by MOMBE.

Figure 2 shows the temperature dependence of the PL spectra observed from the sample A. Strong PL emission in the 1.5-μm-wavelength regions was observed. Although it is usually difficult to observe PL emission from GaAsN layers with the N composition over 1%, a dramatic increase of the PL intensity is observed by the inclusion of Se in the layers. Since the increase of the Se composition in GaAsNSe layers increases the electron concentration,[8] this suggests that their high electron concentrations increase the radiative recombination rate, making the nonradiative recombination in GaAsNSe layers relatively unimportant. Since the electron concentration estimated from a thick GaAsNSe layer was $\sim 8\times 10^{19}$ cm^{-3}, the Fermi energy is estimated to be located at ~ 0.09 eV above the conduction band edge. The broad luminescence with the half width at half maximum of 0.11 ~ 0.14 eV may be caused not only by the degeneracy of the conduction band, but also by the potential fluctuations in the layers due to the increase of the N as well as Se compositions.

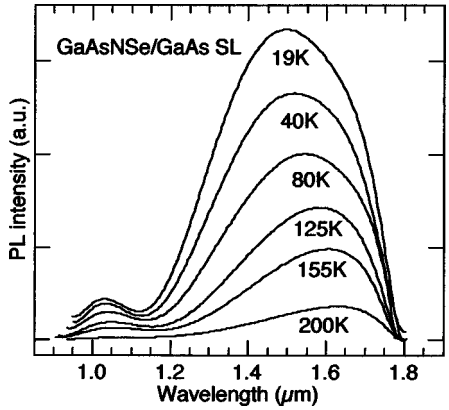

FIG. 2. PL spectra for the GaAsNSe/GaAs SLs (sample A) measured at temperature of 19–200 K.

Figure 3 shows the temperature dependence of the PL peak energy for the sample A (open circles). To check the alloy composition dependence of the PL peak energy, the one for the sample B with the lower N composition is also plotted in Fig. 3 with the closed circles. The N and Se compositions of the sample B are 1.2% and 7.6%, respectively. Upon increasing the N composition in GaAsNSe layers, a redshift of the PL peak energy was observed. In addition, the PL peak energy of the GaAsNSe/GaAs SLs varied only slightly with temperature. The differential temperature coefficients dEg/dT are -3.5×10^{-4} and -2.9×10^{-4} eV/K for the open and closed circles, respectively. These values are close to -3.1×10^{-4} eV/K for the GaAsN alloys with 0.8%–3.8% N compositions estimated near room temperature and are lower than -5×10^{-4} eV/K for GaAs.[9]

These observations of the long-wavelength emission demonstrate the possible applications of the GaAsNSe/GaAs SLs as active layers in long-wavelength light emitters. However, as is shown in Fig. 2, the PL intensity is quenched for the higher temperature. The increase of the N and Se compositions in the GaAsNSe alloys will increase the average tensile strain in GaAsNSe/GaAs SLs due to the smaller lattice constants of GaN and Ga_2Se_3 in the zinc-blende structure.[8] When the N and Se compositions in GaAsNSe are 2.2% and 2.9% in sample A, respectively, the biaxial tensile strain in the GaAsNSe layers will be -5.6×10^{-3}, and the average strain is the biaxial tension on the order of -8.9×10^{-3}. Therefore, to compensate the average tensile strain in the sample, the GaAsNSe/GaAs-SL active layers were

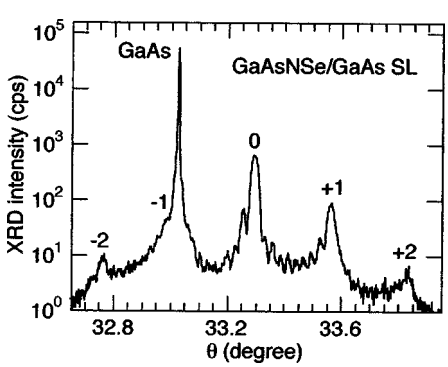

FIG. 1. Typical (004) XRD profile of GaAsNSe/GaAs SL structure grown on GaAs. The N and Se compositions in GaAsNSe are 2.2% and 2.9%, respectively.

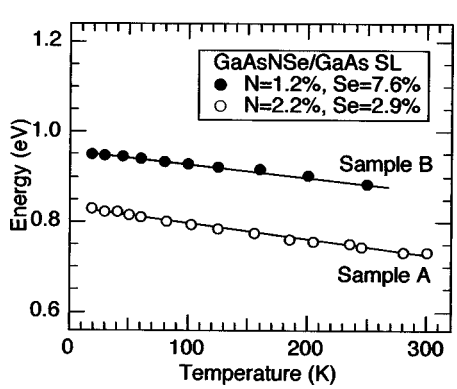

FIG. 3. Temperature dependence of the PL peak energies for the samples A (open circles) and B (closed circles).

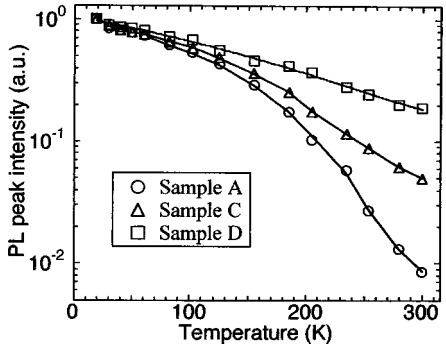

FIG. 4. Temperature dependence of the PL peak intensity for the samples A, C, and D.

sandwiched by GaAsN/GaAsSb-SL layers, which will have biaxial compressive strain of $+7.3\times10^{-3}$. This will also form effective separated confinement heterostructure (SCH) with additional GaAs cladding layers.

Figure 4 shows the temperature dependence of the PL peak intensities for three samples with different structures. The sample A without the strain compensating GaAsN/GaAsSb SLs showed the PL intensity significantly reduced near room temperature as discussed earlier. However, the reduction of the PL peak intensity for the higher temperature was improved in the SCH sample C, and the PL intensity measured at 300 K was $\sim$5% of the intensity measured at 19 K. One factor accounting for this improvement is the reduced average strain, as shown in Table I. In the case of sample D, SL periods were reduced compared with sample C to reduce the total thickness of the strained layers. The temperature dependence of the PL intensity was much reduced, and the PL intensity at 300 K was as large as 20% of that at 19 K. This improvement is attributed to the reduced defect density in each SL by minimizing the strain accumulation in the SLs. The activation energies estimated from the Arrhenius plot of the integrated PL intensity of the samples A, C, and D were 74, 96 and 98 meV, respectively. These results demonstrate that the optical quality can be substantially improved by the elimination of nonradiative defects by minimizing the average strain as well as the strain accumulation in the layers. Since the SCH structures discussed in this letter can be luminescent in the broadband wavelength range of 1.2 to 1.6 μm, this GaAsNSe/GaAs SL will be a candidate with high potentiality to realize broadband SLDs on GaAs substrates.

In conclusion, we reported the optical properties of the GaAsNSe/GaAs SLs. Strong PL emission at around a 1.5-μm wavelength was observed from the as-grown GaAsNSe/GaAs SLs without thermal annealing. This was attributed to the effect of the high electron concentration in the conduction band, which increase the radiative recombination rate. We also discussed the improvement of the optical property from the viewpoint of lattice strain in the samples. The temperature dependence of the PL peak intensity was drastically improved by combining the GaAsN/GaAsSb SLs with the GaAsNSe/GaAs SLs to reduce the strain accumulation in the samples, and bright PL emission was observed up to room temperature. The (GaAsNSe/GaAs SLs)/(GaAsN/GaAsSb SLs)/GaAs SCH and obtained results demonstrate the possibility of fabricating SLD on GaAs substrates for WDM optical-fiber comunications the wavelength region around 1.55 μm.

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