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Formation of Natural InAlAs Vertical Superlattices

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$In_xAl_{1-x}As/In_yAl_{1-y}As$ vertical superlattices (VS) were naturally formed by phase separation during the growth of InAlAs layers at temperatures ranging from 565℃ to 615℃ by metalorganic chemical vapour deposition(MOCVD). The VS lies perpendicular to the (001) growth plane. CuPt-type atomic ordering had occurred in some of the layers. Bandgap reduction of ~300 meV was observed in the InAlAs layers grown at temperatures ranging from 565 ℃ to 615 ℃. Such a large reduction in bandgap energy was attributed to combined effects of the VS and CuPt-type ordering. It is suggested that the vertical superlattice structures may be used as a new technique for the formation of quantum wire structures.

1. Introduction

There is increasing evidence that natural superlattices can be spontaneously formed, either by phase separation or atomic ordering, during the growth of nominally homogeneous epitaxial layers composed of a wide range of ternary III-V alloys.[1] This structural formation is expected to have significant influences on the fundamental properties of the materials such as electronic band structure or free carrier mobility. In general, the bandgap energy of ordered structure is smaller than that of unordered structure, but the difference of two bandgap energies is not more than ~150 meV. In this letter we report (a) the observation of naturally formed $In_xAl_{1-x}As/In_yAl_{1-y}As$ vertical superlattices (VS) in the layers of nominal composition $In_{0.52}Al_{0.48}As$ grown on exactly (001) oriented InP substrates, (b) the occurrence of CuPt-type ordering in some of the layers and (c) a large bandgap reduction of ~300 meV in PL peak energy for the $In_xAl_{1-x}As/In_yAl_{1-y}As$ VS structures grown in the range 565 - 615 ℃.

2. Experimental

The $In_{0.52}Al_{0.48}As$ layers were grown in a low-pressure MOCVD reactor using trimethylindium, trimethylaluminum and arsine as sources. The substrates were Fe-doped semi-insulating (001) InP on which $In_{0.52}Al_{0.48}As$ layers ~500nm thick were grown at a rate of 0.5nm/s with temperatures ranging from 565 ℃ to 800 ℃. Photoluminiscence (PL) measurements were performed at 77K using 1-m double monochromator equipped with liquid N_2 cooled Ge detector and GaAs photomultiplier. For each sample, two orthogonal [110] and [$\bar{1}$10] cross-section and [001] plan-view thin films were prepared by mechanical polishing and Ar+ ion milling using a liquid N_2 cold stage, and examined by transmission electron microscope (TEM) and transmission electron diffraction (TED) in a JEM 2010 instrument operated at 200kV.

3. Results and discussion

[$\bar{1}$10] cross-section (002) TEM dark field (DF) examinations of the layers of nominal composition $In_{0.52}Al_{0.48}As$ grown at 565 ℃, 590 ℃ (Fig. 1(a)) and 615 ℃ show the presence of quasi-periodic vertical band-like contrast with the bands lying perpendicular to the (001) growth plane. The contrast in these images is sensitive to a variation in alloy composition. The bands alternate in contrast from bright to dark, and the bands within each layer correspond to material with two different alloy compositions, indicating that phase separation has occurred. These bands are termed vertical superlattices (VS). For the $In_{0.52}Al_{0.48}As$ layers grown in the range 680 ~ 800 ℃, analogous (002) DF images showed a single uniform contrast because the layers were of uniform composition. [110] cross-section (002) TEM DF image of the $In_{0.52}Al_{0.48}As$ layers grown at 565℃, 590℃ and 615℃ showed uniform contrast, indicating that there is no vertical band in this cross-section. As the growth temperature increased from 565 ℃ to 615 ℃, the VS became less planar and uniform, and decreased in thickness from ~15nm to ~6nm.

In Fig. 1(b) is shown [001] plan-view TEM (020) DF image of $In_{0.52}Al_{0.48}As$ layer grown at 590 ℃. The image shows quasi-periodic band-like contrast with the bands lying parallel to the [$\bar{1}$10] direction. This is in good agreement with the cross-section DF results. Plan-view (002) images from the layers grown at 590 ℃ and 610 ℃ showed similar bands, although the band thickness and uniformity depended on the growth temperature.

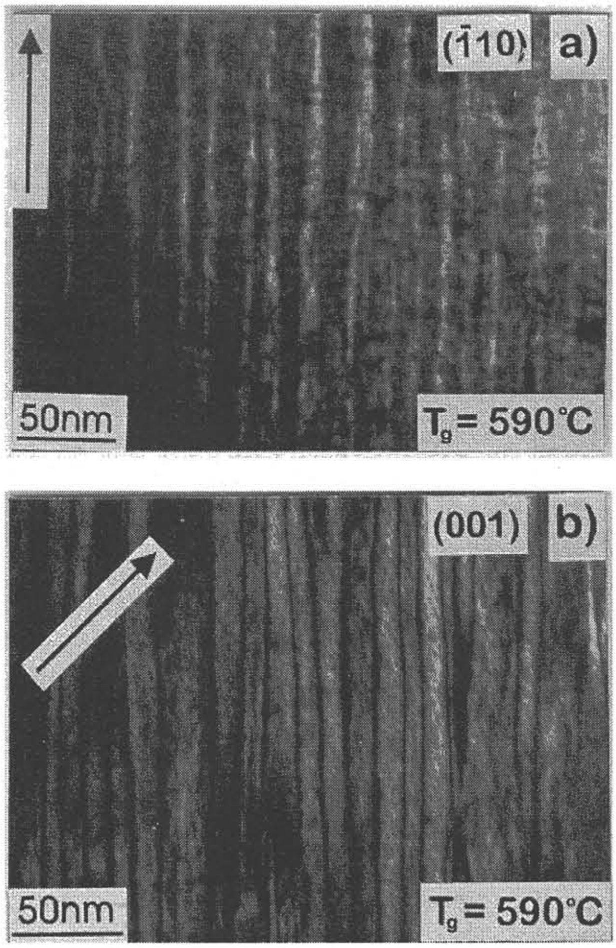

Fig. 1. (a) [1̄10] cross-section (002) TEM dark field (DF) images of $In_{0.52}Al_{0.48}As$ layers grown at 590 ℃ showing quasi-periodic vertical band-like contrasts with the bands lying perpendicular to the (001) growth plane. (b) [001] plan-view (020) DF image of the 590 ℃ layer showing quasi-periodic band-like contrasts with the bands lying along the [1̄10] direction.

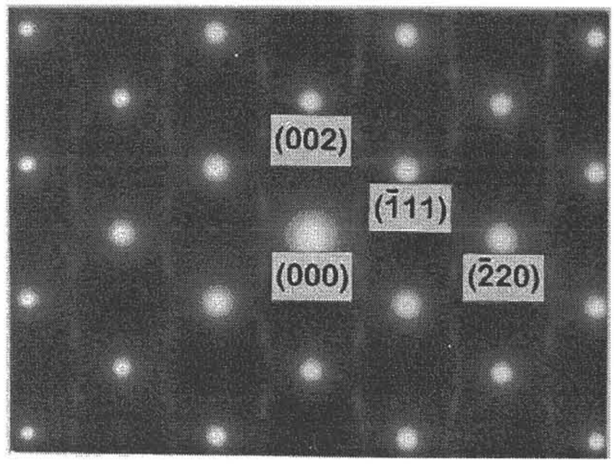

Fig. 2. [110] cross-section TED patterns obtained from In0.52Al0.48As layers
grown at 615*C showing main lattice spots and 1/2{111}superlattice spots.

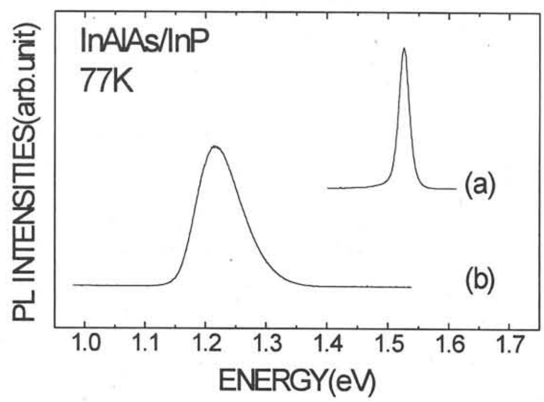

Fig. 3. Normalized PL spectra at 77 K of InAlAs layers grown at 775 ℃ (a), 565 ℃(b).

Fig. 2 shows [110] TED pattern taken from an $In_{0.52}Al_{0.48}As$ layer grown at 615 ℃, which shows the main spots and ½{111} superlattices spots, indicating the presence of CuPt-type ordering on the {111}B planes. The superlattice spots are elongated and tilted $\sim 16^{\circ}$ from the [001] direction. The superlattice spots are joined together by [001] lines of weak diffuse diffracted intensity. The existence of weak diffuse intensity could be attributed to the elongation and inclination of ordered microdomains.[2] [110] TED examinations showed that as the growth temperature increased from 565 ℃ to 800 ℃, the ordering increased, reached a maximum at ~650 ℃ and then decreased. The layers grown at temperatures above 700 ℃ showed no evidence for CuPt-type ordering.

Cheng et al.[3] showed that vertical quantum wells lying along the [001] growth direction formed in MBE (GaAs)n/(InAs)n short-period superlattices (SPS) grown on (001) InP and suggested that the strain induced from the deviation of superlattice periodicity in the SPS could be responsible for the formation of the vertical quantum well. The present InAlAs VS is different from the growth behaviors observed by Cheng et al., since in this work uniform single thick $In_{0.52}Al_{0.48}As$ were grown. Peiro et al[4] investigated MBE InAlAs buffer layers grown on (100) InP vicinal surfaces and reported quantum-wirelike structures corresponding to In-rich and Al-rich materials. No quantum wirelike contrast was, however, observed in the exact (100) substrates. The present VS is also different from the quantum wirelike structure, since in this work the layers were grown on exact (001) InP substrates. Phase separation could be responsible for the VS, though the precise mechanisms are not clear yet.

Normalized 77K PL spectra of InAlAs layers grown at 775 ℃(a) and 565 ℃(b) are depicted in Fig. 3. The PL spectra of the InAlAs layers grown at different growth temperature show single peak with energy position at 1.526 eV and 1.218 eV ,respectively. The peak with energy at 1.526 eV originate from band to band transition of non structured InAlAs alloy matched to InP lattice.[5] But VS structured InAlAs layer grown at 565 ℃ displays red shift as large as ~300 meV. To our best knowledge, this is the biggest red-shift associated with natural superlattices reported so far. It is noted that the 565 ℃ layer showed no CuPt-type ordering. As the growth temperature increased from 565 ℃ to 800 ℃, peak energy monotonically increased and full width at half maximum became narrowed. Gomyo et al.[6] showed that CuPt-type and triple period ordered InAlAs MBE layers grown in the range 520 ~ 570 ℃ experienced a bandgap reduction of ~80 meV. The bandgap reduction in the present InAlAs layers is far greater than that observed by Gomyo et al. Thus this result indicates that a combination of the VS and CuPt-type ordering would produce such a large red-shift.

4. Summary

In summary, we show that InAlAs vertical superlattices were naturally formed by phase separation during the growth of InAlAs/InP layers at temperatures in the range 565~615℃ by metalorganic chemical vapour deposition. The VS structured InAlAs layers including CuPt-type ordering display large bandgap reduction by ~300 meV. The results indicate that the vertical superlattice structures may be used as a new technique for the formation of quantum wire structures.

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