**Optical properties of spontaneous lateral composition modulations in AlAs/InAs
short-period superlattices**

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Abstract

The effect of lateral composition modulation, spontaneously generated during the epitaxial growth of a AlAs/InAs short-period superlattice, on the electronic band structure is investigated using photo-transmission and photoluminescence spectroscopy. Compared with uniform layers of similar average composition, the presence of the composition modulation considerably reduces the band gap energy and produces strongly polarized emission and absorption spectra. We demonstrate that the dominant polarization can selectively be aligned along the $[\bar{1}10]$ or $[010]$ crystallographic directions. In compressively strained samples, the use of (001) InP substrates slightly miscut toward $[111]A$ or $[101]$ resulted in modulation directions along $[110]$ or $[100]$, respectively, and dominant polarizations along a direction orthogonal to the respective composition modulation. Band gap reduction as high as 350 meV and 310 meV are obtained for samples with composition modulation along $[110]$ and $[100]$, respectively. Polarization ratios up to 26 are observed in transmission spectra.

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Lateral composition modulation (CM) is a periodic variation in alloy composition in a direction perpendicular to the growth direction. Lateral CM bears strong similarities with spinodal decomposition observed in bulk alloys, but it also has its own distinct features: since the periodic composition modulation occurs spontaneously, lateral CM falls under the class of self-assembled phenomena, see Figure 1. A one-dimensional CM is characterized by its average composition and the amplitude, wavelength, and direction of the modulation. One method to induce the lateral CM is by growing a vertical short-period superlattice (SPS). CM in SPS structures had been first observed and described in GaP/InP SPS [Hsieh-APL90, Mascarenhas-SM92] and later observed in GaAs/InAs [Cheng-APL92] and AlAs/InAs SPS [Millunchick-MRS97]. These structures show a large band gap reduction and a strong polarization anisotropy with respect to a uniform alloy with a similar average composition. These two effects can be exploited for device design. In principle, the band gap can be decreased without increasing the lattice-mismatch of the epitaxial layer with its substrate. In addition, the polarization dependence of the transition probability can be used in the design of polarization-sensitive devices. CM in GaP/InP has been successfully applied for device applications [Chou-APL95].

A comprehensive understanding of the formation of lateral composition modulation in strained SPS has not yet been achieved. The spontaneous formation of the modulation is likely to result from concurrent effects related to growth, thermodynamics, strain, and surface kinetics. A very good review on this subject is given in Ref. [MRS-Boston99].

The periodic sequence of In- and Al-rich regions creates a lateral superlattice and results in carrier confinement in the In-rich regions. Compared with the band gap of an

alloy with a similar average composition but without modulation, the lowest-energy confined state of the superlattice can be significantly lower. Along with the band gap reduction, the transition probability becomes polarization dependent if measured along the growth direction. The absorption and emission associated with the lowest-energy transition is polarized along the direction perpendicular to the CM direction. The polarization ratio could, in principle, reach factors of more than 100 [Zhang-PRB98]. The band gap reduction and polarization anisotropy depend on the characteristics of the modulation and the band gap difference between the two binary compounds used. Of the three alloy systems mentioned above, AlInAs is the one with the highest band gap difference between the two binary compounds, therefore it is expected to show the largest band gap reduction [Zhang-PRB98]. Indeed, strong band gap reductions were reported [Jones-TSF99]. These results on CM AlAs/InAs were based on low-temperature photoluminescence (PL) results. As will be demonstrated, estimation of the band gap reduction from PL can be misleading since the experimental spectra are often dominated by transitions originating below the gap or from localized regions with strong deviations from the average composition.

In this work we report photo-transmission (PT) and PL measurements on samples showing strong CM along [110] or [100]. The band gap reduction is unambiguously determined by absorption measurements. The control over the direction of the dominant polarization of the emission/absorption process can be achieved by controlling the CM direction. A preferential CM direction was selected by growing the structures on slightly misoriented substrates.

The CM was induced by growing a SPS by molecular beam epitaxy. The samples consist of 100-period $(\text{AlAs})_m\text{-(InAs)}_n$ superlattice grown on a 0.1 μm thick $\text{Al}_{0.48}\text{In}_{0.52}\text{As}$ buffer layer lattice matched to the InP (001) substrate. m and n represent the number of monolayers grown for each component of the superlattice. For the work presented here, we used two samples grown under global compression. They are labeled using their respective direction of the CM, i.e., CM [110] and CM [100]. The values for (n,m) were (1.4,2.0) and (1.5,1.9), respectively. These samples were extensively characterized by techniques such as x-ray diffraction (XRD) [Lee-JMS99], transmission electron microscopy [Norman-APL98,Norman-MRS99, Follstead-APL00], and atomic force microscopy. As determined by XRD, the average In composition of the SPS and the modulation wavelength were 58.5% and 252 Å for CM [110] and 54.8% and 329 Å for CM [100]. XRD can also be used to estimate the relative strength of the modulation [Lee-JMS99]. The normalized integrated intensity of diffraction satellites along the modulation direction is respectively 2.31 and 3.37 for CM[110] and CM[100], indicating a 46% stronger modulation for CM[100].

PT was measured using a conventional set-up by normalizing the change in transmission ΔT upon carrier injection by the absolute transmission T . The frequency-modulated transmission was induced with a frequency-doubled YAG laser (532 nm) and was detected with an InGaAs photo-diode. The PT is approximately related to photo-absorption by $\Delta T/T = -d \Delta\alpha$, where d is the sample thickness.

When grown on on axis (001) substrates, SPS-induced CM often occurs simultaneously in more than one direction, yielding a two-dimensional modulation [Norman-APL98, Norman-MRS99]. As seen from TEM, these samples exhibit relatively

short wire like structure in the growth plane. A slight misorientation of the substrate favors the modulation aligned closer to the tilt direction. In contrast to the 2-D modulation observed for samples grown on on axis substrates, the wires can be predominantly oriented along a unique direction and their average length can be increased significantly. This 1-D modulation is advantageous since they usually have better defined electronic properties. CM[110] and CM[100] were grown on substrates with a 2° miscut toward [111]A and [110], respectively.

Figure 1 shows the TEM (002) dark-field plan-view images for these two samples. The (002) pole is selected for its sensitivity to composition changes and the relatively low contribution of lateral strain to the image contrast. Clear and dark regions are associated to an In-rich and Al-rich alloys, respectively. Cross-section micrographs showed that this modulation is strong already after the growth of ~ 20 periods of the SPS and extends quite uniformly to the sample surface, see Ref. [Norman-MRS99] for more details.

Figure 2 shows PT spectra measured at 77K on CM[110] along and perpendicular to the CM direction. The PT signal shows a clear maximum at 1.023 eV. The energy of this transition, produced by the lateral superlattice, corresponds to a band gap reduction of 0.350 eV compared to a uniform alloy with the same average composition compressively strained to InP. The inset shows the polarization dependence of the PT measured at 1.023 eV. The signal is strongly polarized in the $[\bar{1}10]$ direction and weakly polarized parallel to the CM. The measured maximum ratio of polarizations is equal to 26. Polarized PL spectra measured at 77K revealed a similar transition located approximately 15 meV lower (see arrow in Fig.2). Even though the polarization ratio

measured from PL was only 3.9, the PT and PL features are likely to share the same origin. The absolute amplitude of the composition modulation is not known, it is therefore difficult to compare the experimental results with theoretical calculations. However, it is possible to estimate the amplitude of the modulation from the band gap reduction. Using the theory developed by Zhang et al. [Zhang-PRB98] and taking into account the In rich average composition of the SPS, a band gap reduction of 350 meV corresponds to a modulation amplitude of approximately 13 %. As predicted by Zhang et al. [Zhang-PRB98], for given modulation characteristics, the band gap reduction can be larger for AlAs/InAs SPS than for InAs/GaAs (~ 100 meV) and GaP/InP SPS (~ 300 meV).

Figure 3 shows PT and PL spectra measured at 77K on the second sample, CM[100]. The PT shows a polarized feature at 1.156 eV. This energy is associated with the band gap of the lateral superlattice. The PL spectra measured at 77K has a single feature located at 1.045 eV. PL yields a transition significantly lower in energy than the signal observed from PT. Low-temperature PL spectra can be dominated by radiative transitions below the band gap, originating from defects or from local deviations of any of the modulation characteristics, and therefore do not provide an accurate estimate of the band gap energy. The inset of Figure 3 shows the polarization dependence of the PT measured at 1.156 eV. The angle of maximum intensity is tilted by 12° towards [110]. Transmission electron diffraction revealed that the net CM direction is slightly tilted by approximately 10° towards the same direction. The direction of dominant polarization is very sensitive to the orientation of the CM. The band gap reduction observed in this sample is 310 meV. Compared to CM[110], the relative quality of CM[100], as judged

from the strength of the optical signal, is inferior. Using only PL to estimate the band gap reduction would have led to a much higher, but erroneous, value.

The band gap reduction is higher for CM[110] even though the modulation amplitude for CM[100] estimated from XRD is higher. Assuming that the difference in modulation wavelength has a relatively small effect [Mattila-PRB99], it is possible that the orientation of the modulation has a relatively important effect on the band gap of the structure. No theoretical calculations have been published to date for a [100] modulation.

In summary, we have reported the band gap reduction, the polarization direction, and the polarization ratio from AlAs/InAs short-period superlattices showing a strong lateral composition modulation. The modulation was induced by the growth of a vertical short-period superlattice. In these compressively strained samples, the modulation direction could be selected with the choice of the right substrate miscut.

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FIGURE CAPTIONS.

Figure 1: TEM (200) dark field micrographs of a) CM[110] and b) CM[100].

Figure 2: Photo-transmission (77K) measured on CM [110]. The inset shows the PT intensity as a function of the polarization direction measured at a energy of 1.0231 eV

Figure 3: Photo-transmission and photoluminescence measured at 77K on CM[110]. The solid and dotted curves represents the [010] and [100] polarizations, respectively. The inset shows the PT intensity as a function of the polarization direction measured at 1.156 eV.

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