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Lateral Composition Modulation Induced Structural Anisotropy in InP/GaInP Quantum Dot System

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1. Introduction

Self-assembled quantum dots (SADs) are compressive strained nanocrystals in a dome-like shape usually prepared by Stranski-Krastanov growth of semiconductor materials such as InGaAs/GaAs and InP/GaInP.[1,2] Since the process is simple, the dots are coherently organized and free of structural defects at the initial stage of the three-dimensional (3D) island formation, SADs are becoming more and more interesting for both fundamental research and device applications.

During the dot formation, various intrinsic processes are involved, for example, partial strain relaxation, compositional intermixing and segregation, anisotropic surface diffusion as well as long-range ordering of the ternary materials. These processes greatly modify the dot structures and optical properties. One of the interesting phenomena with potential application would be the polarized photon emission from the quantum dots through structural anisotropy by lifting the degeneracies of the geometrical symmetries. Up to now, such structural anisotropy has only been investigated based on the elongation of the quantum dot shape.[3,4] The use of external anisotropy of the matrix to break the symmetry of the quantum dots, for example, by fabricating the quantum dots inside a quantum wire, a vertical quantum well or superlattice has not been investigated. Because GaInP lattice matched on GaAs intrinsically possesses Cu-Pt type long-range ordering[5-7] and in some case, lateral compositional modulation,[7,8] it is ideal to use InP/GaInP system to investigate the influence of anisotropic matrix to the physical properties of the quantum dots, which is the focus of this study.

2. Experimental

InP/GaInP quantum dot samples were prepared in a gas-source molecular beam epitaxy (GS-MBE). Gallium, indium and aluminum were used as the group III sources while AsH_3 and PH_3 cracked at 950°C were used as group V sources. The substrates were GaAs (001) vicinal surfaces inclined 1° toward the $[1\bar{1}0]$ direction. Details on the growth procedure will be reported elsewhere. After growth of a GaAs buffer, a 140nm GaInP/4ML InP/140nm GaInP dot structure was grown at 480°C. For comparison, samples were also grown at 500°C and 520°C, respectively. During the growth, reflection high-energy electron diffraction showed a 2x1 reconstruction on the GaInP surface. After the growth, X-ray diffraction showed that the lattice mismatch between the GaAs and GaInP is less than 5×10^{-4}. Cross-sectional TEM investigations were performed on a JEM-2000EX TEM operated at 200kV. PL measurements were performed at 2K with an Ar^+ laser as the excitation source and a charge coupled device (CCD) camera as the detector. In order to study the polarization in the PL spectra, a polarizer was inserted before the light was collected into a fiber to the monochromater.

3. Results and Discussion

Structural anisotropy in InP/GaInP SADs.

Three kinds of structural anisotropy in the InP/GaInP quantum dot system were examined. They are ellipsoidal base shape of the InP island with its long axis in the $[1\bar{1}0]$ direction, lateral composition modulation of GaInP along the [110] direction and the Cu-Pt_B type long-range ordering in the GaInP matrix.

Fig. 1 shows the representative (110) and $(1\bar{1}0)$ cross-sectional bright field images of the InP/GaInP dot structures grown at 480°C. InP dots have an ellipsoidal base elongated in the $[1\bar{1}0]$ direction. The average base length along the [110] and $[1\bar{1}0]$ direction is 35nm and 45nm, respectively. The average dot height is 5.5nm. In addition, lateral composition modulation in GaInP was observed in the $(1\bar{1}0)$ cross-section, which has been known to occur when growing $(GaP)_2/(InP)_2$ super-lattice on GaAs (001) by GS-MBE [8]. Indium and gallium-rich domains extended in the $[1\bar{1}0]$ direction were formed alternatively along the [110] direction. Such lateral composition modulation is enhanced at the GaInP/GaAs interface and over the InP dots, suggesting a strain effect nature of the phase separation in the GaInP.

Selective area transmission electron diffraction (TED) pattern of InP/GaInP showed wavy streaks of the superspots at $(\frac{\bar{1}}{2}, \frac{1}{2}, \frac{1}{2})$ and $(\frac{1}{2}, \frac{\bar{1}}{2}, \frac{1}{2})$ and other equivalent positions along the [110] zone-axis. It indicates the presence of two long-range ordering variants of Cu-Pt_B type in the $[\bar{1}11]$ and $[1\bar{1}1]$ directions in GaInP. In both TEDs along the [110] and $[1\bar{1}0]$ zone-axis, small super-streaks beside the high-order diffraction spots along the [001] axis in the reciprocal

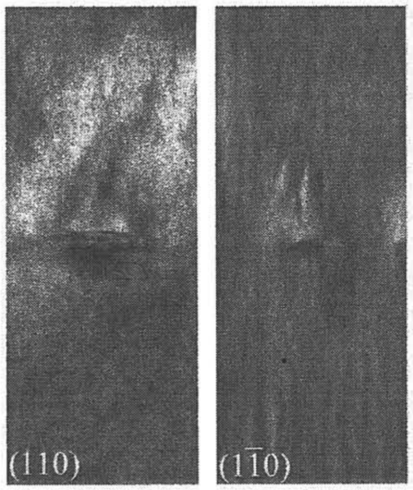

Fig.1. Cross-sectional TEM images of InP/GaInP SADs. Lateral composition modulation is observed in the (1$\bar{1}$0) cross-section, or along the [110] direction in GaInP.

space were observed. These streaks are contributed by the large lattice InP dots. This indicates that the InP dot lattice differs from the GaInP lattice mainly in the [001] direction. To our knowledge, this is the first experimental report showing the lattice distortion in the embedded InP/GaInP SADs.

Polarized PL from InP/GaInP SADs

Polarized PL emissions from ordered GaInP materials have been well known in literature.[6,7] Due to the presence of Cu-Pt$_B$ type ordering, PL from the (001) GaInP surface showed polarized PL in the [110] direction. In the present study, however, the PL spectra from both GaInP matrix and InP dots are polarized in the [1$\bar{1}$0] direction, as shown in **Fig. 2**. The degree of polarization from the InP dots $(I_{//}-I_{\perp})/(I_{//}+I_{\perp})$ is 43% while that from the GaInP is even larger. Such a result suggests that a mechanism opposite to the long-range ordering is taking the role. In fact, due to the nature of MBE growth, surface diffusion lengths for the Ga and In atoms in the [1$\bar{1}$0] direction on the (001) surface are longer compared to that in the [110] direction, which is just opposite to that of MOVPE growth[9]. As shown in **Fig. 1**, such anisotropic surface diffusion induces not only elongated InP island shape but also lateral composition modulation during the growth of GaInP. The modulation is strong at a growth temperature of 480°C or less, and is reduced at higher than 500°C. Such a lateral modulated structure behaves like a vertical superlattice. Referring the work of InAs/GaAs SADs, size elongation usually gives weak polarization (~15%)[4]. The major mechanism that gives rise to the polarized PL in this study is believed to be the lateral composition modulation in GaInP matrix. Lateral composition modulation in the GaInP not only gives rise to a polarized PL from the matrix, but also a polarized PL from the InP dots.

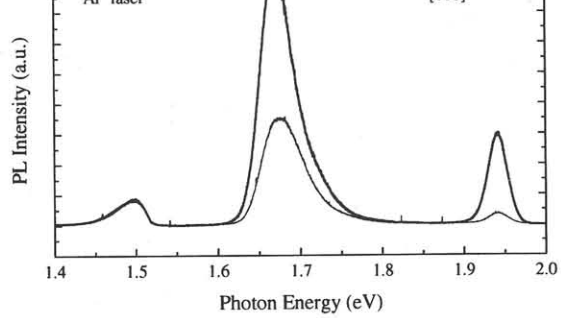

Fig. 2. 2K PL of InP/GaInP SADs. Spectra from both the InP dots and the GaInP matrix are polarized.

4. Conclusions

Based on the above study, it is concluded that a strong optical anisotropy in the [1$\bar{1}$0] direction of InP/GaInP SADs system grown by GS-MBE at low temperature is contributed mainly by the lateral composition modulation in the GaInP matrix.

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