MOVPE GROWTH OF SMALL AND UNIFORM INP/GAINP QUANTUM DOTS

HONG-WEN REN, MITSURU SUGISAKI, JEONG-SIK LEE AND SHIGEO SUGOU

Single Quantum Dot Project, ERATO, JST, c/o Optoelectronics and High Frequency Device Research Labs. NEC Corporation, 34 Miyukigaoka, Tsukuba, 305-8501, Japan
E-mail: ren@obl.cl.nec.co.jp

YASUAKI MASUMOTO

Single Quantum Dot Project, ERATO, JST, and Institute of Physics, University of Tsukuba, Tsukuba, 305-8571, Japan

InP self-assembled quantum dots embedded in $Ga_{0.5}In_{0.5}P$ were fabricated by metal-organic vapor phase epitaxy. Small size and highly uniform InP dots were obtained at lower growth temperature by using tertiarybutyl-phosphine instead of phosphine. Growth of 4ML InP on a 4ML GaP spacer at 550^{o}C resulted in InP islands of about 30nm in base diameter and 7nm in height within 10% deviation. The formation of the dots was sensitively monitored by a fast-nulling ellipsometer. By inserting another 2ML GaP spacer between the InP dots and the GaInP cap-layer, the half width of the 2K PL spectra from the InP dots was reduced to 30meV. The anomalous spectra from the InP wetting layer and undeveloped clusters were removed by alloying them with the GaP spacers.

Recent years there is much interest in the self-assembled growth of semiconductor quantum dots by Stranski-Kranstanow (S-K) mode because these dots can be defect-free and are of high optical quality.[1-4] However, due to the random initiation of the 3-dimensional (3D) islands, controlling on their spatial and size distribution has been a tough issue limiting their potential applications. In the case of metal-organic vapor phase epitaxy (MOVPE) of InP/GaInP SADs, even though the bimodal size distribution for the coherent islands can be overcome by using a GaP spacer, the island size is still large and the areal density of dots is low.[2,3] Since large dots may introduce dislocations and low density leads to poor optical efficiency, growth of small size, high density and uniformity InP dots becomes imperative.

It is well known that the S-K grown 3D-island size reduces with the growth temperature. However, in MOVPE growth of InP/GaInP SADs using PH_3, the decomposition temperature of PH_3, which is 650^{o}C for 50% decomposition, limits the reduction of the growth temperature. Since the decomposition temperature for tertiarybutyl-phosphine (TBP) is much lower (475^{o}C for 50% decomposition) than that for PH_3, we investigated the growth and optical properties of small size and highly uniform InP/GaInP SADs on the GaAs (100) substrates by using TBP in a MOVPE system.

Growths were carried out in an EMCORE D-75 MOVPE system on the semi-insulating GaAs (001) substrates. The growth pressure was 60 torr at a H_2 flow of 12 standard litres per minute and substrate rotation is 1400 revolutions per minute. Triethylgallium (TEGa), trimethylindium (TMIn), AsH_3, PH_3, TBP were used as the growth sources. A fast-nulling ellipsometer was used to monitor the growth process with a 632.8nm laser source at 70^o incidence. After growth of a GaAs buffer, 4 monolayers (ML) InP dot layer sandwiched between 250 nm $Ga_{0.5}In_{0.5}P$ barriers was grown. The growth temperature was between 530^oC and 660^oC. The mole fractions of TEGa, TMIn and PH_3 for the growth of GaInP were about 1.6×10^{-5}, 1.55×10^{-5} and 1.5×10^{-2}, respectively. During the growth of InP dots, TBP with a mole fraction of 1×10^{-3} was used to replace PH_3. The growth rates for GaInP and InP were 0.8 ML/s and 0.5 ML/s, respectively. A Nanoscope IIIa atomic force microscopy (AFM) and a JEM-2000EX transmission electron microscopy (TEM) were used to investigate the dot structures. Photoluminescence (PL) spectroscopy was performed at 2K and 77K with an Ar^+ laser at 514.5 nm as the excitation source.

At the temperature range from 530^oC to 660^oC, growth of lattice-matched GaInP on GaAs produces a surface with wide terraces and small size monolayer islands. Direct growth of InP dots on such a surface results in bimodal distribution of the dot size, and growth of 2~4ML GaP spacer can help reduce the number of undeveloped islands.[2,3] We found the GaP spacer produced a [110] elongated mound modulated surface with the average mound width, length and height to be about 30nm, 120nm and 3~4ML, respectively. When 4ML InP was deposited on the GaP modulated surface at higher than 540^oC, well-developed InP islands with high size uniformity were obtained as shown in **Fig. 1**. The island size increases with the growth temperature as **Fig. 1a** and **1b** show the InP islands grown at 550^oC and 600^oC, respectively. In both cases, the size deviations are less than 10%.

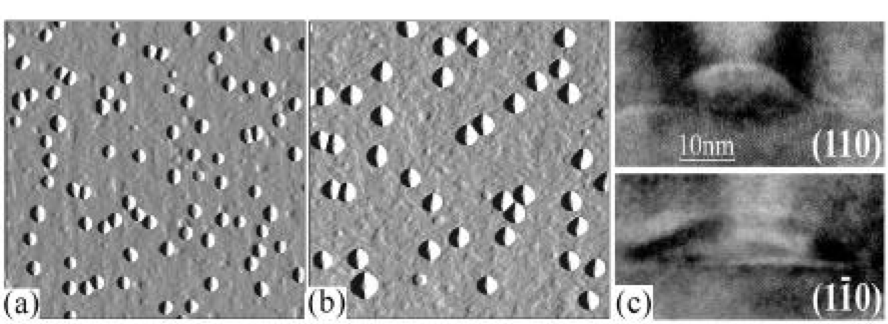

Figure 1. AFM images (1μm×1μm) of 4ML InP islands grown on 4ML GaP/GaInP at **(a)** 550^oC and **(b)** 600^oC, **(c)** TEM images of 550^oC grown InP dots in the (110) and $(1\bar{1}0)$ cross-sections.

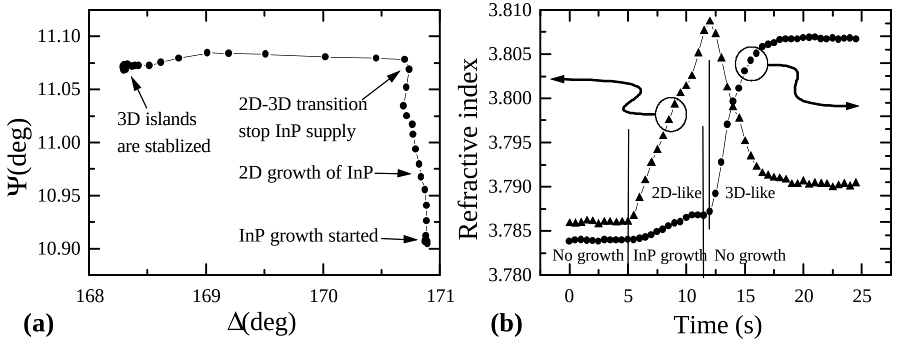

Figure 2. The Δ–Ψ trajectory **(a)** and change of *n, k* **(b)** during the growth of InP dots showing the 2D-3D transition by fast-nulling ellipsometer. The sampling time is 25ms and data interval is 0.5s.

No 3D-islands were formed with 4ML InP below 540°C. After embedding the InP islands grown at 550°C with GaInP, the typical sizes of the InP dots were 7nm, 25nm and 35nm along the [001], [110] and $[1\bar{1}0]$ directions, respectively (**Fig. 1c**).

The fast-nulling ellipsometer is found powerful in monitoring the dot formation and improving the controllability. A spiral in the Δ–Ψ trajectory is obtained during the growth of GaInP on GaAs. When InP was deposited, as shown in **Fig. 2a**, the growth began with a 2D nature where a steady increase in Ψ and minor change in Δ was observed. When more than 3.8ML InP was deposited, a drastic decrease in Δ with little change in Ψ was detected. Analyses reveals that the decrease in Δ corresponds to the increase in the surface extinction coefficient *k* due to 3D surface roughening, as shown in **Fig. 2b**. The overshooting in the refractive index *n* reveals strain accumulation and relaxation in the surface layer. The critical thickness for the 2D to 3D transition was thus determined to be 3.8ML and was also confirmed *ex-situ* by AFM. With increasing the growth temperature for the InP dot formation, the critical thickness reduces slightly while the extinction coefficient increases enormously, displaying that the island size increases with the growth temperature.

Finally the optical properties of InP dots was studied. The temperatures for growing InP and GaInP were 550°C and 580°C, respectively. When 4ML InP was grown directly on GaInP, broad PL spectra from the dots were observed at 1.652eV with a FWHM of 55meV, as shown in **Fig. 3a**. When a 4ML GaP spacer was added before InP growth, the FWHM was reduced to 35meV due to the improved uniformity of dot sizes (**Fig. 3b**). By inserting another 2ML GaP interface layer prior to the growth of the GaInP cap-layer, the FWHM was further reduced to 30 meV as shown in **Fig. 3c**. The use of GaP spacers also raised broad spectra from the undeveloped InP clusters at around 1.76-1.86eV. This means that although the

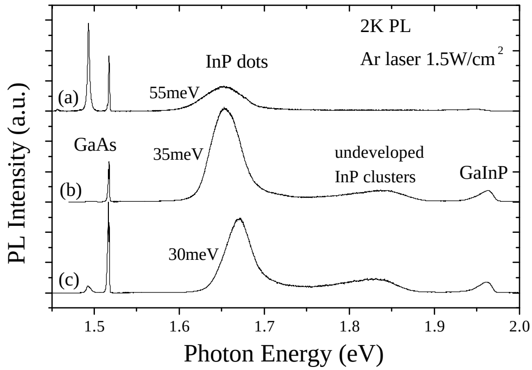

Figure 3. 2K PL of InP/GaInP dots **(a)** with no GaP spacer, **(b)** with a 4ML GaP spacer below the InP dots, **(c)** with a 4ML GaP spacer below and a 2ML GaP spacer above the InP dots.

underlying GaP spacer improved the size uniformity of the developed InP islands, there remain thin InP clusters probably in the depressions of the GaP modulated interface. Since these thin InP clusters are sandwiched by GaP spacers, an easy way to remove them is to alloy them with the GaP layers during the subsequent growth. By growing the GaInP cap-layer at a temperature higher than 630°C, these anomalous spectra were removed and the FWHM of PL from the InP dots is as small as 29meV at both 2K and 77K.

In summary, highly uniform and small size InP/GaInP SADs were prepared by MOVPE using TBP at 550°C. The growth of a 4ML GaP spacer layer before growing InP SADs improved the dot size uniformity and the insertion of another 2ML GaP spacer before growing the GaInP cap-layer further reduced the PL FWHM from the InP dots to 30meV or less.

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