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Self-organized InAs quantum dots formation by As/P exchange reaction on (001) InP substrate

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In this letter, we present the results of InAs quantum dots (QDs) prepared on a (001) InP substrate. As/P exchange reaction at the surface of InP buffer was used to form the InAs islands in the reactor of low pressure metalorganic chemical vapor deposition at 600 °C. Preliminary characterizations of the InAs QDs have been investigated by using atomic force microscopy and photoluminescence (PL). Room temperature PL emission from the 0-dimensional system centers at 1520 nm and the full width at half maximum of the PL is 92 meV. © *1998 American Institute of Physics.* [S0003-6951(98)01919-6]

Three-dimensional (3D) confinement of carriers has received much attention for quantum device applications as well as for fundamental study in quantum physics.[1] In order to fabricate the structures showing the predicted quantum effects, various techniques such as wet or dry etching, ion beam implantation or milling, or regrowth on processed samples have been investigated.[2–5] However, these methods result in large nonradiative recombination of carriers by damage or impurities at the surface or the interface.

As an *in situ* fabrication technique, Stranski-Krastanov (S-K) growth on lattice-mismatched substrates by molecular beam epitaxy (MBE) or metalorganic chemical vapor deposition (MOCVD) has been recently recognized as a promising way to eliminate such nonradiative recombination. Moreover, many reports have been presented to fabricate the quantum dot (QD) structures by using this technique.[6–10]

The exchange of group V atoms on the surface of a III-V semiconductor, due to their important function in the growth of high quality quantum well heterostructures, has been studied by many authors.[11–14] Direct evidence of the As–P exchange reaction on the surface of (001) InP under MOCVD conditions has been observed by using surface photoabsorption (SPA), and it has been demonstrated that the reaction can occur above 360 °C.[12] The reaction under MBE conditions has also been studied by using reflection high energy electron diffraction (RHEED)[13] and by reflectance anisotropy spectroscopy (RAS) recently.[14] The exposure of the InP surface to As flux (or AsH_3) results in the formation of a thin pseudomorphic InAs layer on the InP surface.[11–15] The structural properties of such an InAs layer have been investigated in some detail.[15] However, the previous reports have not considered the effect of strain on the InAs layer, which will result in the formation of InAs islands during As/P exchange reaction. In fact, the islanding of the InAs layer plays an important role in the As/P exchange reaction. In this letter, we present the results of the formation of the InAs QDs on the InP substrate by using As/P exchange reaction. Preliminary characterizations of the InAs QDs have also been performed by using atomic force microscopy (AFM) and photoluminescence (PL) spectra.

The samples used here were prepared on a (001) InP substrate by low pressure MOCVD with a horizontal quartz reactor. 100% arsine (AsH_3), phosphine (PH_3), and trimethylinium (TMIn) were used as source materials. The carry gas was Pd-purified H_2. Total H_2 flow rate was 6 ℓ/min. The pressure of the reactor was kept at 76 Torr. After the substrate treating at 650 °C under PH_3 for 5 min, the InP buffer layer with 200 nm was grown at 600 °C, and then the sample was exposed to AsH_3 at the same temperature with different period of 2, 7, 10, and 30 s, respectively, which correspond to samples A, B, C, and D. As/P exchange reaction occurs on the InP surface during this stage to form InAs materials. Subsequently, the InP cap layer with 50 nm was grown immediately. To investigate a morphology of the InAs islands formed during As/P exchange reaction, sample E without the InP cap layer, but with reduced temperature under AsH_3 from 600 to 350 °C, was performed. In all procedures the flow rate of TMIn (17 °C), PH_3, and AsH_3 were 4.4×10^{-6}, 2.0×10^{-3}, and 4.4×10^{-4} mol/min, respectively.

The AFM measurements were performed by using Nanoscope III (Digital Instruments). Photoluminescence measurements were performed at room and 10 K temperature by using a 632.8 nm line of He–Ne laser. The average excitation density was about 300 mW/cm^2.

Figure 1(a) shows the AFM image of sample E. One can see clearly in Fig. 1(a) that many large islands are formed by the As/P exchange reaction at 600 °C as well as the process of reducing the temperature. The mean size of the islands is about 2000 nm, and the island shape is irregular. The lattice mismatch between the InAs and InP is over 3%. Hence, the formation of the InAs islands, like the S-K growth mode, is driven by a strain of the InAs layer. However, the InAs layer formed here is due to As/P exchange reaction rather than deposition. The exchange reaction is easily saturated if the reaction occurs homogeneously on the surface. However, owing to the formation of islands, the layer in some regions is thin enough that the reaction is not saturated and is continuous. Hence, although the time of the As/P exchange reaction at 600 °C is only 10 s, the time of reducing the sam-

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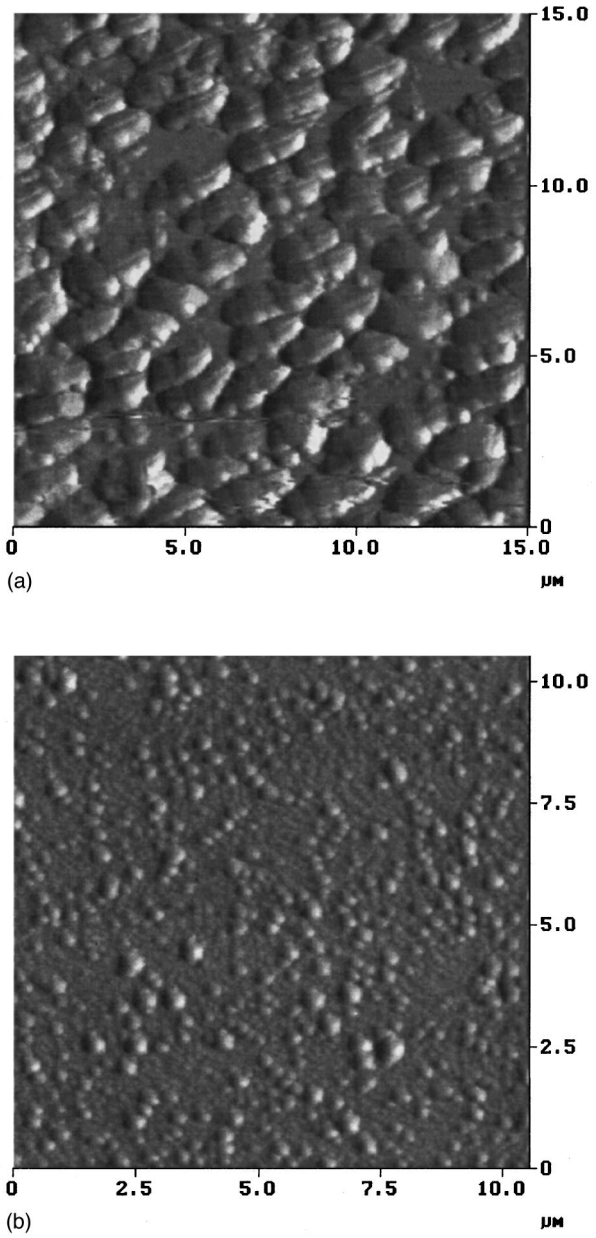

FIG. 1. The AFM image of the samples without the InP caplayer: (a) After As/P exchange reaction of 10 s at 600 °C, the sample's temperature was cooled down under AsH_3. (b) After deposition of the InAs with nominal 3 MLs at 490 °C, the sample's temperature was cooled down under AsH_3.

ple's temperature is very long (about 10 min from 600 to 350 °C). The reaction during the stage of reducing the temperature makes the InAs islands very large. Figure 1(b) shows the AFM image of another sample that InAs with nominal 3 monolayers (MLs) was deposited on the InP buffer at 490 °C, and then its temperature was lowered under AsH_3. The InAs islands (about 200 nm wide) as well as many small islands are seen clearly, and the island shape is round. As/P exchange reaction during the stage of reducing the temperature still increases the InAs islands size. However, compared with sample E, the size of the islands is much smaller even though the InAs with nominal 3 MLs was deposited on the InP surface before reducing the temperature. It is obvious that the amount of InAs in sample E is more than that in the sample shown in Fig. 1(b). The results suggest that: (1) As/P exchange reaction at higher temperature is relatively stronger; (2) the reaction can be controlled by varying the reaction time and reaction temperature; (3) the reaction still occurs at least in the coverage of nominal 3 MLs InAs.

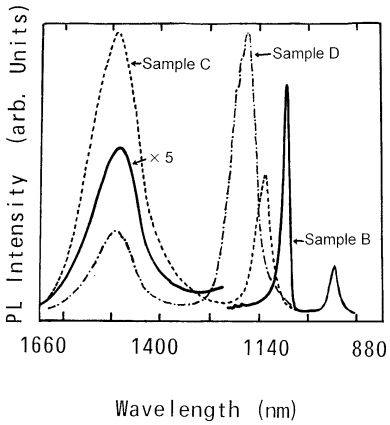

FIG. 2. The PL spectra of the InAs QDs of samples B, C, and D measured at 10 K temperature, which were formed by using As/P exchange reaction on the InP surface.

Figure 2 shows the 10 K PL spectra of samples B, C, and D, respectively. For sample B as shown in the Fig. 2, three peaks at 925, 1020, and 1440 nm are seen clearly. They can be attributed to the InP bulk, the InAs wetting layer (WL), which corresponds to the effects of InAs/InP strained quantum well (SQW), and the InAs QDs, respectively. In this method, like the S-K growth mode, the InAs islands also were formed on the top of two-dimensional (2D) InAs WL. Because the density of the InAs islands is not high enough, the photogenerated carriers cannot be wholly captured by InAs QDs. The recombination of some carriers in the 2D InAs WL can be also observed at the lower temperature. The full width half maximum (FWHM) of the PL is about 23 meV, and it is almost the same as that of the InAs/InP SQW grown by MOCVD.[16] Other samples without the InP cap layer or the process of As/P exchange reaction, but with other preparing conditions being constant, were also measured at 10 K temperature. However, any signal has not been taken except for the peak at 925 nm. It confirms that the peaks at 1440 nm and around 1020 nm are due to the As/P exchange reaction.

Sample C (shown in the Fig. 2) also shows 1440 and 1060 nm peaks with emission from the QDs and WL respectively. We have noted that the peak of the QDs is much stronger than that of sample B. It indicates that the InAs island density of sample C is higher than that of sample B. With increasing island density, more and more photogenerated carriers relax from a 2D WL into zero-dimensional QD levels, which results in a decrease of the PL intensity and of the WL and an increase of the PL intensity of the QDs. However, the QD PL intensity of sample D is lower than that of sample C. It suggests that the bigger incoherent islands were formed due to the longer reaction time (30 s) which results in nonradiative recombination of carriers. On the other hand, the peak at 1116 nm (1.111 eV), corresponding to 4 MLs InAs WL,[16] was much broaden and two shoulders appear on the lower energy side. This indicates that the thickness of the InAs layer is not uniform and there is a grade of composition in the InAs/InP interface. In addition,

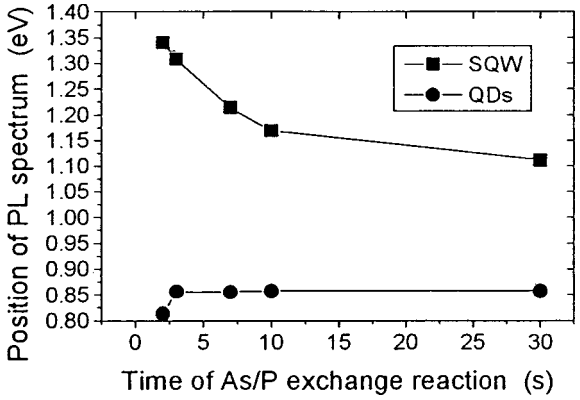

FIG. 3. The position of 10 K PL spectra as function of the As/P exchange reaction time: (■) InAs wetting layer; (●) InAs quantum dots.

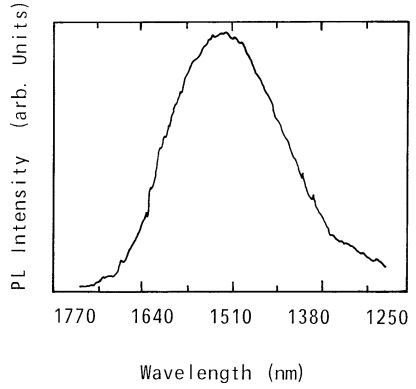

FIG. 4. The room temperature PL spectrum of sample C.

for the PL spectra of sample A (not shown here), there was a very weak peak around 1440 nm, which indicates that the density of InAs islands is very low due to the short time of anion exchange reaction. However, the peak centered at 934 nm (1.327 eV) is strong and narrow, which can be attributed to InAs/InP SQW with 1 ML InAs layer.[16]

Figure 3 shows an energetic position of the PL peak emission from the InAs WL and QDs as a function of the reaction time. One can see clearly in Fig. 3 that the positions of the QD PL peaks are similar except for sample A, whose reaction time is 2 s. This might indicate that the lateral size of islands formed in the initial stage is large, and the island density is very low as discussed previously. With the increase of reaction time in the initial stage, the island size will become small, and the density increases rapidly, which results in an increase of the PL intensity. However, by further increasing the reaction time, which corresponds to the increasing amount of InAs, the islands will become large again, and some incoherent islands are formed, resulting in a decrease of the PL intensity. The change process of the islands agrees with our experiment's results. Similar results have also been observed by using *in situ* AFM measurements while depositing InAs on GaAs substrate under MBE conditions.[17] In addition, although the islands formed in the initial stage will change with increased reaction time, the formation of more new islands makes the distribution of the island size unchanged. It results in similar positions of the QD PL. In Fig. 3, we also found that the thickness of the WL increases with reaction time. However, the rate of increase in the thickness will decrease with the increase in reaction time. It suggests that the rate of As/P exchange reaction decreases with the increase in reaction time due to the influence of the thickness InAs WL.

Figure 4 shows a room temperature PL spectrum of sample C. Strong PL emission from InAs/InP QDs is observed, which shows good features of InAs QDs formed on InP by As/P exchange reaction. In addition, the emission from InAs WL was not found, which suggests that most of the photogenerated carriers have been captured by InAs QDs at room temperature. As shown in Fig. 4, the PL peak centers at 1520 nm, and the FMWH is 92 meV.

In conclusion, As/P exchange reaction on the surface of InP, as a promising way, can be used to *in situ* fabricate the InAs self organized QDs. In our preparation, the strongest PL spectrum emission from the InAs QDs was obtained from sample C, in which the time of the As/P exchange reaction was 10 s. The room temperature PL spectrum centers at 1520, and the FMWH of the PL is about 92 meV. In addition, the PL measured at 10 K temperature suggests also the emission from the WL. When the reaction time varies from 2 to 30 s, the peak positions change in the region of 1.327–1.111 eV. The narrowest peak was obtained from sample B with 7 s of As/P exchange reaction.

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