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InAs SELF-ASSEMBLED NANOSTRUCTURES GROWN ON InP(001)*

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The 6–period stacked layers of self-assembled InAs quasi-quantum wires(qQWRs) and quantum dots(QDs) embedded into InAlAs on InP(001) substrates have been prepared by solid molecular beam epitaxy. The structures are characterized by atomic force microscopy(AFM) and transmission electron microscopy(TEM). From AFM we have observed for the first time that InAs qQWRs and QDs coexist, and we explained this phenomenon from the view of the energy related to the islands. Cross-sectional TEM shows that InAs qQWRs are vertically aligned every other layer along the growth direction [001], which disagrees with conventional vertical self-alignment of InAs QDs on GaAs substrate.

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Recently, the growth of low-dimensional semiconductor materials has attracted considerable interest for their great potential in optoelectronic and detector devices.[1,2] For example, quantum dots(QDs) and quantum wires(QWRs) lasers are predicted to have such characteristics as lower threshold current, wider modulation bandwidths, and better temperature stability[2,3], compared with quantum wells(QWs). Conventionally, various fabrication methods of QDs and QWRs have been proposed based on high resolution patterning of two-dimensional(2D) heterostructures by combining wet and dry etching. This process could produce good results[4]; however, it has some fatal drawbacks such as its complexity, the limited resolution of electron lithography, the defects created by the ion etching process. So, in order to fabricate high quality QDs and QWRs, a novel technique called Stranski-Kranstanow growth mode(S-K) has been used as a route for the fabrication of defect-free In(Ga)As QDs on GaAs[5], and InAs QWRs on InP.[6]

The spontaneous formation of small coherent islands on a few wetting layers via S-K growth mode was driven by the reduction of the total strain and surface energy of the layer-substrate systems. At the same time, it is limited by kinetic factors, such as atomic diffusion etc. There were a lot of theories about the formation and evolution of the islands.[7,8] Various Ⅲ-Ⅴ semiconductor alloys, such as In(Ga)As/GaAs[5], InP/InGaP[9], InAlAs/AlGaAs[10], InAs/InP[11], with pronounced preferential elongation of QDs or QWRs under specific growth conditions, have been fabricated by molecular beam epitaxy(MBE) or metal-oxide vapor phase epitaxy(MOVPE). To gain more uniformity of nanostructures, vertical self-alignment of QDs along the growth direction during the subsequent deposition of multilayer of islands separated by thin spacer layers has been introduced experimentally[12] and theoretically[13], such as InAs QDs on GaAs substrates. But only a few reports are available regarding the vertical alignment InAs/InP.[14]

InP substrate is another very important semiconductor material other than GaAs. If we choose suitable highly strained epitaxy materials, for example, by using the binary InAs as the low band-gap semiconductor, then we can obtain new light-emitting devices in the 1.3–1.5 μm wavelength range; this is very important for optical fiber communication systems. The lattice mismatch between InP and InAs is 3.2%, which is twice smaller than that of the well-known InAs and GaAs system(the lattice mismatch is 7%), so some different phenomena, for example, the islands ordering along [1$\bar{1}$0] direction[11], could be observed by depositing a few monolayers of InAs on InP substrate. In this letter, we report on the formation of elongated QWRs in the 6-period InAs/InAlAs heterostructures on InP (001) substrate. Atomic force microscopy (AFM) and transmission electron microscopy (TEM) observations revealed a large concentration of elongated QDs, which we called quasi-QWRS(qQWRs) with the peculiar characteristics of vertical self-alignment every other layer.

The samples studied in this work were grown on semi-insulating InP (001) substrates by a Riber 32p Molecular Beam Epitaxy at 500 ℃. After thermal desorption of the oxide on InP, a 0.2 μm In-

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AlAs was grown as buffer layer at a growth rate of 0.7 μm/h. The lattice mismatch of the InAlAs buffer layer on InP substrates was determined to be less than 10^{-3} by *ex situ* double-crystal X-ray diffraction. Following 10 s growth interruption, an InAs layer was deposited at 0.4 ML/s. Sample A contains 6 layers of 6.5 ML InAs, separated by 30 nm InAlAs without capped layer. TEM observations revealed that sample B was composed of 6 layers of 6 ML InAs layer, separated by 10 nm InAlAs, and then a 40 nm thick InAlAs cap layer was deposited. During the growth, substrate rotation and an As_4 pressure of 2.7×10^{-4} Pa were used. A transition from a streaky to a spotty pattern during InAs deposition was observed by *in situ* reflection high-energy electron diffraction (RHEED), suggesting that InAs growth mode changing from 2D to 3D. The transition occurs approximately between 2 and 2.5 ML, which disagrees with the result for $In_{0.5}Ga_{0.5}As/GaAs$[10] (its lattice mismatch is slightly larger than InAs/InP). In the present case 2.1 nm InAs was required for the spontaneous formation of islands. This suggested that InAs on InAlAs/InP has an equilibrium different from In(Ga)As on GaAs in surface, interface, bonds, and dislocation energies.[15] The growth temperature was monitored using a substrate thermocouple. Finally, the sample was cooled under arsenic pressure. AFM measurements were performed using a Digital Instrument Nanoscope Ⅲ in contact mode (with force of about 0.58 N/m). Sample B was studied by TEM at 300 kV accelerated voltage in conventional way.

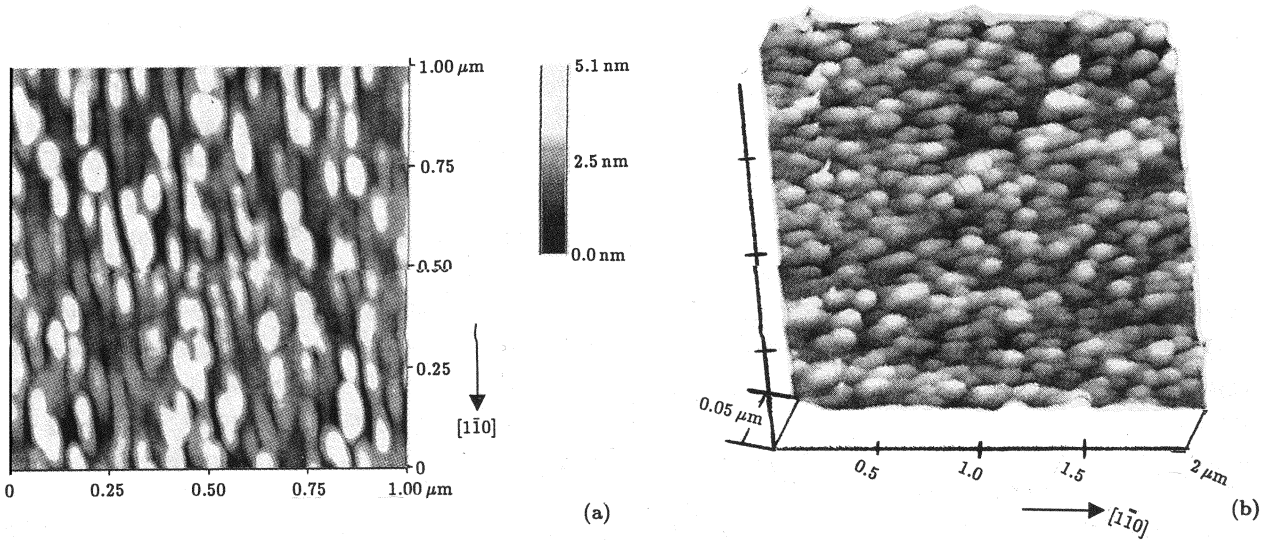


Fig.1. AFM image of uncapped InAs nanostructures. (a) Top view, (b) 3D pattern.

Figure 1 shows typical AFM images of the surface morphology and 3D uncapped InAs layers. It is clearly observed that there are two kinds of nanostructures. One is well-separated islands apparently like ellipsoids whose dimensions are approximately 110 nm long, 60 nm wide, and 5 nm high. The other is 3D nanostructures appearing as closely packed wires, whose dimensions are about a few hundred nanometer long, 60 nm wide, and 3–5 nm high, elongated in the [1$\bar{1}$0] direction. This observation agrees with the measurements made by Shen[16], who demonstrated that the QWRs are pseudomorphic along the line direction and the relaxation of strain occurs normal to the wire and in the growth direction revealed by X-ray diffraction.

These results (measured by AFM) clearly demonstrate that InAs qQWRs and QDs are formed simultaneously under our growth conditions. According to the theoretical mode[8,10] on understanding the characteristics of QD based on the consideration of island nucleation and strain energy, we can get some insight into the cause of the coexistence of QDs and qQWRs. Nucleation is governed by random surface process: steps, defects, or local thickness fluctuations. All islands rapidly reach a certain size, below which they are unstable, and the size and shape of the islands correspond more and less to an optimum of elastic relaxation.[17] Asynchronous nucleation and fast initial growth rate combined are proven to be the major origin of the size fluctuations of InAs QDs. Although the formation of islands relieves most of the strain energy and consequently retards the introduction of

misfit defects, they also build up some strain with further deposition via adatoms diffusion and/or island coarsening. When the stress is big enough, under the conditions of high density of QDs, it seems that some coherent big islands coalesce along the $[1\bar{1}0]$ direction in order to relax more stress[6], thus form qQWRs. To facilitate our explanation above, it needs further confirmation by TEM. But it is true that once islands coalesce, a large amount of misfit strain is reduced. In other words, the incoherent qQWRs are more preferential sinks for adatoms than coherent ones. The coherent islands grow little or not. This explains why the qQWRs coexist with coherent QDs.

Figure 2 shows the (200) dark field cross-sectional TEM micrograph of multiple qQWRs along $[1\bar{1}0]$ and light spots are observed. Since the (200) structure factor is very sensitive to the local composition but not lateral strain, we attribute the light spots to InAs. On viewing along $[1\bar{1}0]$, the light spots appear as white stripes, which is consistent with the micrograph observed by AFM. From Fig 2, we can observe that the

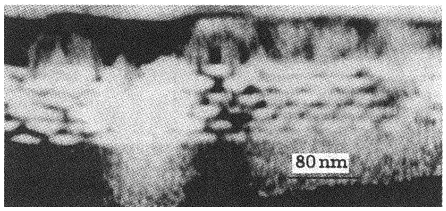


Fig.2. TEM image of cross-section 200 dark field of InAs nanostructures.

spots become bigger and bigger from the first layer to the sixth layer and some spots join with neighboring ones in the top layers. This is consistent with the transition observed in RHEED from streak to spotty, which occurs earlier in the top layers than in the bottom layers. This indicates that critical thickness for planar growth in stacked layers is reduced and the excess material due to a thinner wetting layer accumulates to form large qQWRs.[18]

Apart from the characteristic mentioned above, we have observed another interesting and the most important phenomenon: these light spots are vertically aligned every other layer in the growth direction, not like InAs/GaAs[13] in vertical self-assembled multiple sets of islands, in which the QDs of the first layer are reproduced in the upper layers. The difference between the two kinds of vertical alignment is, we think, due to the different strains. For the multilayer In(Ga)As QDs on GaAs substrate, the buried islands produce a tensile stress that reduces the lattice mismatch of InAs in the spacer (GaAs) above the islands. The In adatoms forming the next layer will preferentially migrate from the region of higher to lower lattice mismatch and accumulate on the top of lower islands. For the multilayer InAs qQWRs on InP, the tensile stress caused by buried qQWRs exists between qQWRs, not above qQWRs, so in adatoms favor sites between qQWRs. By using the finite element technique Wu[14] revealed that the most far reaching direction for elastic distortion is in a plane, the one making an angle of about 30° with the normal to the elasticity-extended line in the material. But he did not take into account the influence of the neighboring buried qQWRs, and the InAlAs alloy ordering. We think that there will be a more complex mechanism for it and further work is in progress.

In summary, we have demonstrated that in the multilayer of InAs nanostructures in InAlAs/InP, there exist some peculiar characteristics: InAs QDs and qQWRs coexist. In addition, the vertical alignment occurs every other layer, which is different from conventional vertical self-organized alignment of InAs QDs on GaAs substrate.

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