

# Epitaxial growth and optical properties of ultra-low density InAs quantum dots on patterned substrate by MBE

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In this paper, we had investigated site-controlled and low-density quantum dots(QDs) required communications band single-photon source on the patterned substrates by molecular beam epitaxy (MBE). There were still difficulties in isolating single QDs from the low-density QDs assemble. In order to overcome such difficulty, growth of site-controlled QDs on pre-patterned substrates were proposed and preserve high-material quality of the low-density QDs for a real single photon, the In(Ga)As/GaAs QDs grown on the high reflectivity distributed bragg reflector mirror composed of a limited thickness of the wavelength of the micro-cavity center. The spectrum measured at 10K, and the wavelength was 1.3156 $\mu\text{m}$ , width only was 45 $\mu\text{eV}$ , respectively. The results showed that the site-controlled and low-density QDs growth technology would be prepared not only had high optical quality, but also for communications band 1.3 $\mu\text{m}$  single-photon emission.

(Received November 13, 2014; accepted January 21, 2015)

*Keywords:* Site-controlled, Low-density, Quantum dots, Single-photon, MBE

## 1. Introduction

In recent years, scientific investigation and its technological applications at the nanometer scale have gained an increasing interest in the research community. Due to unique three-dimensional quantum confinement and their potential device applications, semiconductor quantum dots have attracted considerable interests in the last three decades. Using Stranski-Krastanow growth method, low threshold, high thermal stability and high quantum efficiency lasers, and other active components have been demonstrated in In(Ga)As/GaAs QDs systems [1, 2]. More recently, it was also demonstrated that QDs can be efficiently used as single-photon emitters for quantum cryptography applications [1-4] and growth efforts have been devoted to the fabrication of low-density QDs samples to perform single dot spectroscopy. It was demonstrated that a uniform distribution of low-density InAs QDs emitting at 1.3  $\mu\text{m}$  at 10 K with high efficiency can be obtained by employing a combination of ultra-low In(Ga)As/GaAs growth rate and InGaAs capping layer [2-4]. Clean exciton-biexciton dynamics, anti-bunching, and electroluminescence from single QD were observed, making these QDs promising candidates for single-photon sources at 1.3  $\mu\text{m}$  [5-11].

In this paper, we reported the optical properties and growth of site-controlled and ultra-low density InAs/(In)GaAs QDs on the patterned substrate, the high material-quality of the low-density QDs for a real single-photon growth by MBE, the spectrum measured at 10K, the wavelength was 1.3156 $\mu\text{m}$ , width only was 45 $\mu\text{eV}$ . The results show that the site- and low-density layer quantum dots prepared not only has high optical

quality, but also for communications band 1.3 $\mu\text{m}$  single-photon emission.

## 2. Experiment details

The samples were prepared by MBE on (111)B GaAs patterned substrates. They are GaAs (or InGaAs) capped low-density InAs QDs embedded in GaAs matrix. The (In)GaAs capping layer has a layer thickness of 5nm and an In composition of 15%. The samples were grown by the method described in Ref. 5 and 6. The growth rates of low-density QDs were 0.002 and 0.1 monolayer/s, respectively.

The patterning procedure of substrate was as follow. Firstly, a rigid SiO<sub>2</sub> layer and a scanned electron beam resist (PMMA) were deposited on the (111)B GaAs wafer in order to benefit from the crystallography in the etching and growth steps. After development, the mask was transferred to the SiO<sub>2</sub> layer using a dry-etched by means of a BCl<sub>3</sub>/N<sub>2</sub> inductively coupled plasma (ICP) etching, and the resist was removed by acetone ultrasound bath and oxygen plasma. HF etching is finally used to remove the remaining SiO<sub>2</sub> and the native oxide prior to growth.

These samples were respectively characterized by PL measurements, which was excited with a 632.8nm He-Ne laser and monitored by an un-cooled InGaAs detector.

## 3. Results and discussion

Fig. 1 shows the SEM top-view image of a substrate patterned with 100 nm-side after etching. The lateral InAs/(In)GaAs QDs were not shown since they were very

thin as compared to the other structures. Morphological characterization of patterned substrates was performed before the growth. In particular, the prepared holes will cause the effective growth rate to increase dramatically in the patterned substrate. The In adatoms were more sensitive to capillarity because of their longer diffusion length. This causes an anisotropic deposition of the InAs layer. QDs forms at the whole apex, which presents the strongest local surface (concave) curvature. QDs structures were also deposited in the whole edges of the patterned. The QDs grown on the sidewalls were fragile and no associated emission was observed. The SEM images were regularly acquired to check the quality of the dry etching and to measure the whole edges size.

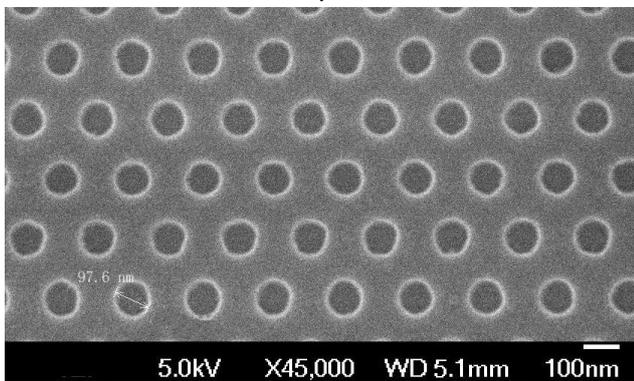


Fig. 1. SEM of top-view image of a patterned substrate.

Before the overgrowth processing, the patterned surface must be cleaned. Fig. 2 shows Ga-assisted defoliation appears to be a viable alternative to the more conventionally used H-assisted defoliation for in situ damage-free removal of the oxide from patterned substrates. If the hydrogen could not be carefully controlled, the H-assisted defoliation will lead to degradation of the surface and Fermi-level surface pinning.

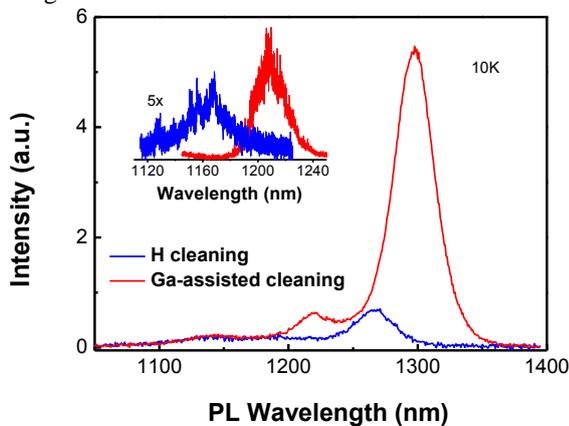


Fig. 2. The PL spectra of the InAs/(In)GaAs QDs grown on the patterned substrate. Inset is high-resolution micro-PL spectra measured at very low excitation power at 10K.

In contrast, the Ga-assisted deoxidization is to the pre-patterned the substrate that it landed itself easily and necessity to integration and remove the surface oxide without damaging the pattern before overgrowth could proceed, and further device processing. The method of oxide removal the gallium, in the absence and in-situ of As, was used to reduce the stable surface oxide to a volatile form, by the reaction  $\text{Ga}_2\text{O}_3 + 4\text{Ga} \rightarrow 3\text{Ga}_2\text{O}$ . This practical advantage deoxidization method can be carried out in the growth chamber with no need for additional apparatus and without causing significant damage to the fine pattern features. By such patterns to control the nucleation site of InAs/(In)GaAs QDs grown by MBE. The PL intensity of InAs/(In)GaAs QDs grown on patterned substrate by Ga-assisted deoxidization is 10 times than H-assisted deoxidization. Inset is high-resolution micro-PL spectra measured at very low excitation power at 10K.

Encouraging progress has been made towards the realization of a single-photon source based on integrating self-assembled quantum dots, though an early proposal for an electrical single-photon source was too weak to allow the second-order correlation function to be studied. An improved emission aperture single-photon, which incorporates an optical cavity formed between the semiconductor-air interface and high-reflectivity Bragg mirror in the aperture. The Cross-section of SEM with the structure forms a pyramidal cavity, which the leaky modes are affected by the cavity, and enhances the efficiency of measurement collection compared with devices without a cavity. The experimentally determined photon-collection efficiency, which is a more pertinent parameter for applications, is typically around 10% [2], because of that not all the cavity mode can be coupled into an experiment and that there is scattering of the mode by the rough pillar edges [2]. It is expected that the photon-collection efficiency could increase with improvements to the new designs or processing technology of micro-cavity, so as to form a lateral variation in the refractive index, which creates a forbidden energy gap for photonic modes in which light can't propagate. Photons could be trapped in a central irregularity in this structure. Fig. 3 shows high-quality active cavities have also been demonstrated in (In)GaAs containing InAs quantum dots.

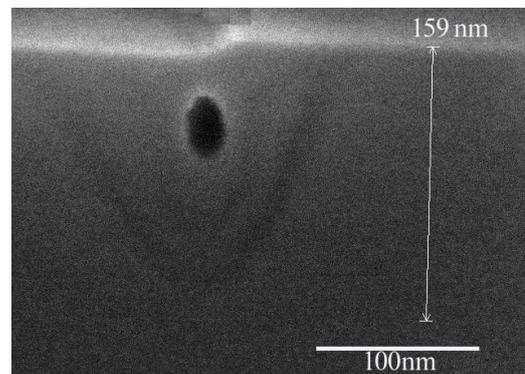


Fig. 3. High-quality active cavities in (In)GaAs containing InAs quantum dots.

Fig. 4 for comparison structures of InAs/(In)GaAs QDs with buried layer GaAs and micro-cavity. Not like their name, InAs/(In)GaAs QDs do not mean that they consist of pure InAs. In fact, they represent regions that are locally rich in indium. Owing to the larger QDs size and higher indium composition of the low-density QDs, typical PL spectra of the InAs/(In)GaAs QDs samples. Spectra of the samples are also shown, which are normalized. For clarity, the spectra are grouped and offset. As compared to the micro-cavity samples, PL peak energy intensity from samples are observed. The energy intensity by micro-cavity are attributed to the In/Ga atomic inter-diffusion across the interfaces of the QDs and their surrounding matrix [7-9]. It was argued that InAs capping layer relaxes the strain in the QDs due to reduced lattice-mismatch between QDs and (In)GaAs capping layer. In(Ga)As/GaAs QDs are very sensitive to QDs and micro-cavity quality, which has been confirmed in the fabrication of single photon emitters. In order to preserve high material quality of the low-density QDs, it was suggested that, in a real single photon device structure growth, the layers on top of the QDs are better to be grown with a temperature not higher than that for the QDs growth.

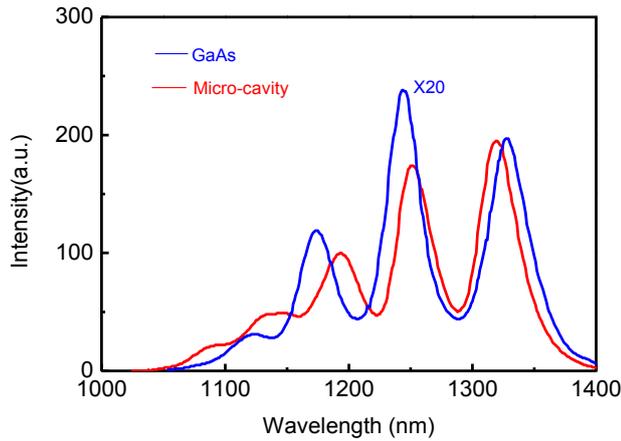


Fig. 4. The comparison of the InAs QDs of PL spectra of buried layer GaAs and micro-cavity at 10 K

Fig. 5 shows poisson statistics can also be used for describing the QDs population for pulsed excitation, but in this case the probability of occupation only describes the initial condition after the laser pulse. The spectrum measured at 10K, the wavelength of 1.3156 $\mu$ m, width only 45 $\mu$ eV, respectively. As shown in Fig. 3.5 where we compare the integrated PL intensities of the spectral lines marked X and BX as a function of excitation intensity for the two-pump regimes. At low- excitation power, the PL intensity dependence on the laser power P for both pump modes can be fitted by the relation  $I_{x,Bx} \propto P^n$ , with  $n = 0.70$  and  $1.35$  for X and BX lines, respectively. The fact that the ratio of the exponents is equal to 2 ( $n_{Bx}/n_x = 2$ ) confirms that the BX line corresponds to the biexciton

emission. The observations not only confirm the site-controlled high optical quality and low-density of the QDs, but also suggest their promising further application to 1.3  $\mu$ m single-photon emission.

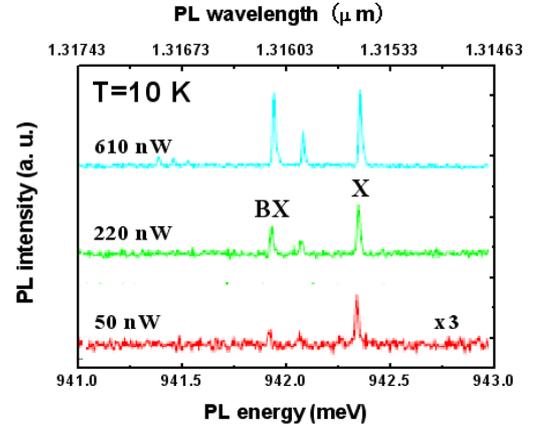


Fig. 5. The spectra of single photon measured with function of excitation intensity for the two pump regimes at 10K

#### 4. Conclusions

In conclusion, Using MBE growth of InAs QDs by site-controlled and low-density In(Ga)As/GaAs QDs has been investigated. The optical properties of the QDs were very sensitive to quality of pre-patterned substrates and QDs' growth quality. In order to preserve high material quality of the low-density QDs for a real single photon, the InAs QDs layer grown on the high reflectivity distributed Bragg reflector mirror composed of a limited thickness of the wavelength of the micro-cavity center. The spectrum measured at 10K, and the wavelength of 1.3156 $\mu$ m, width only 45 $\mu$ eV, respectively. The results showed that the site-controlled and low-density quantum dots growth technology prepared not only has high optical quality, but also for communications band 1.3 $\mu$ m single-photon emission.

#### Acknowledgments

This work was financially supported by the National Natural Science Foundation of China under Grant Nos 61370043,60976056,61308051 and 61006039,NSAF Nos U133306,Science Foundation of JiLin Province (20140520139JH), and Excellent Youth Doctor Foundation of Jilin Agricultural University(No.201411) .

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