Fabrication of In$_{0.15}$Ga$_{0.85}$As Nanohole Templates on GaAs (001) for Quantum Dot Molecules

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1. Introduction
Quantum-dot (QD) molecules, often referred to as artificial molecules, have been an intense field of research for quantum computing [1] and quantum cryptography [2] applications. Commonly, the nanohole template is applied as a key technique for obtaining QD molecules through patterned nanoholes. Several approaches using artificial substrate process methods, such as atomic force microscopy (AFM) tip oxide, and scanning tunneling probe-assisted nanolithography, have been developed to obtain nanohole templates for growing QD molecules [3-5]. These methods, however, require complicated and expensive substrate processing equipment and are also prone to defects and contamination. Recently, Salamo et al. [6] has developed the droplet epitaxy technique to fabricate GaAs nanoholes on GaAs surface. The droplet epitaxy technique is simple, flexible, and without requirement for artificial substrate processing. For the droplet epitaxy growth, only group III element molecular beam is initially supplied to the substrate surface to form group III nanodroplets. The substrate with group III droplet is subsequently exposed to the group V elements beam to crystallize the droplets into III-V nanocrystals [7]. Our group has developed nanoholes to fabricate InAs quadra-quantum dots (QQDs) formation which will be useful for quantum cellular automata applications (QCA). In application-wise, uniform QQDs are needed. Therefore, nanohole templates with isotropic configuration will be key nanostructure providing uniform QQDs. In this work, we report the formation of In$_{0.15}$Ga$_{0.85}$As nanohole templates on GaAs (001) substrates by droplet epitaxy using molecular beam epitaxy. Atomic force microscopy (AFM) measurement was performed to characterize the nanostructure properties. The effects of substrate temperature are studied.

![Fig. 1](image-url) The Evolution of 20 ML In$_{0.15}$Ga$_{0.85}$As nanohole templates to the QQDs: (a) 20 ML In$_{0.15}$Ga$_{0.85}$As nanoholes at 390 °C, (b) 20 ML In$_{0.15}$Ga$_{0.85}$As nanoholes at 450 °C, (c) 1.7 ML InAs QQDs on 20 ML In$_{0.15}$Ga$_{0.85}$As nanoholes at 450 °C.

2. Experiment and results
In this work, all the samples are grown on GaAs (001) substrates in RIBER 32P solid-source molecular beam epitaxy (MBE) system. Prior to the growth, surface oxide desorption is carried out under As$_4$ flux at a beam equivalent pressure of 8×10$^{-6}$ Torr by slowly ramping up substrate temperature until the reflection high energy electron diffraction (RHEED) showed a clearly abrupt pattern. A 300-nm-GaAs buffer layer is then grown at 600 °C with a growth rate of 0.5 monolayer/second (ML/s). Next, the substrate temperature is lowered to the desired temperature without As$_4$ beam to minimize excess As on the surface. Before indium and gallium deposition, the background pressure of the growth chamber is reduced to less than 10$^{-9}$ Torr to minimize the initial interaction between In$_{0.15}$Ga$_{0.85}$As and arsenic during droplet formation. The In$_{0.15}$Ga$_{0.85}$As amount of 20 ML (an equivalent amount of In$_{0.15}$Ga$_{0.85}$As grown on GaAs) is deposited with a constant deposition rate of 1 ML/s at various substrate temperatures of 300 °C, 330 °C, 360 °C and 390 °C. Then, the In$_{0.15}$Ga$_{0.85}$As droplets are exposed to As$_4$ beam for 5 minutes at substrate temperatures of 200 °C to crystallize the nanodroplets. Finally the samples are quenched and taken out from MBE system. The surface morphology of the In$_{0.15}$Ga$_{0.85}$As nanohole template is examined by atomic force microscope (AFM).

Fig. 2(a) shows an AFM image of In$_{0.15}$Ga$_{0.85}$As nanoholes after crystallization. The image is taken from a sample deposited at 360 °C. The sample surface clearly displays the ring-and-hole structure. Cross-section profiles of a typical nanohole structure (area indicated in Fig. 2 (a)) are shown in Fig. 2(b). The outer dimension (o), the inner dimension (i), the depth (d), and the lobe height (h) of the nanohole structure are defined as in Fig. 2(b). For the profile along [100] direction, the nanohole has an outer dimension of $o \approx 173.83$ nm, an inner dimension of $i \approx 68.72$ nm, and a depth of $d \approx 4.09$ nm. For the profiles along [1-10] direction, the nanohole has an outer dimension of $o \approx 186.74$ nm, an inner dimension of $i \approx 66.39$ nm, a depth of $d \approx 4.29$ nm, and density of $7.04 \times 10^8$ cm$^{-2}$. The hole exhibits a V-shape profile along [110] direction and a U-shape profile along [1-10]. The nanohole depth profiles along both directions are different due to anisotropic properties of the crystal. Meanwhile, the nanoholes have an anisotropic lobe structure around the hole with an average lobe height of $h \approx 2.59$ nm along [110] direction and an average lobe height of $h \approx 5.45$ nm along [1-10] direction. From the top-view, the nanohole displays a ring-and-hole structure of circular edge and rectangular hole. The densities of nano-
holes prepared by varying the substrate temperature are shown by black solid line with square symbol in Fig. 3. The densities of nanoholes are $14.2 \times 10^8$ cm$^{-2}$ at 300 °C, $9.24 \times 10^8$ cm$^{-2}$ at 330 °C, $7.04 \times 10^8$ cm$^{-2}$ at 360 °C and $4.8 \times 10^8$ cm$^{-2}$ at 390 °C, respectively. The average outer diameters along [110] direction of nanoholes are 143.83 nm, 169.58 nm, 181.9 nm, and 207.59 nm for the 300 °C, 330 °C, 360 °C and 390 °C samples, respectively; for as shown by black dash line with circle symbol in Fig. 3. The average inner diameters are 69.19 nm, 73.75 nm, 78.72 nm and 78.78 nm, respectively as shown by black dot line with triangle symbol in Fig. 3. The average depths of nanohole are 5 nm at 300 °C, 4.69 nm at 330 °C, 5.05 nm at 360 °C, and 3.79 nm at 390 °C as shown by gray solid line with pentagon symbol in Fig. 3. The nanohole inner dimension distribution with various substrate temperatures is shown by histograms in Fig. 4. The standard deviations of inner dimensions (σ) are 9.38 nm at 300 °C, 10.1 nm at 330 °C, 7.74 nm at 360 °C, and 8.42 nm at 390 °C.

Fig. 2 (a) AFM image of In$_{0.15}$Ga$_{0.85}$As nanohole template formed on GaAs and (b) line profiles of a nanohole template, defined by the dashed line in (a).

Fig. 3 The dependence of average density, outer dimension, inner dimension and depth of nanoholes along [110] direction on deposition temperature.

![Fig. 3](image)

3. Conclusion
Droplet epitaxy is conducted to create nanohole templates. At higher substrate temperature during droplet epitaxy, nanohole templates become bigger in dimension but less number of nanoholes density due to the initial droplet size and density. However, the depth of nanoholes remains nearly the same.

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