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## Fabrication of single- or double-row aligned self-assembled quantum dots by utilizing SiO<sub>2</sub>-patterned vicinal (001) GaAs substrates

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We investigated the formation of In<sub>0.8</sub>Ga<sub>0.2</sub>As self-assembled quantum dots (SAQDs) grown on SiO<sub>2</sub>-patterned 1°, 2°, and 5°-off (001) GaAs substrates by selective area metalorganic vapor phase epitaxy technique. The SiO<sub>2</sub> patterns were filled with various stripe opening windows along the misorientation direction of the substrates. During the growth of the GaAs buffer layer on the opening regions, the steps on the (001) top facet was affected by the widths of the (001) top facet and the misorientation angles of the substrates. Single- or double-row aligned In<sub>0.8</sub>Ga<sub>0.2</sub>As SAQDs having definite interval were successfully fabricated on the (001) top facet with optimized top width and periodicity of step bunching. These results indicate that the selective growth technique of SAQDs by utilizing SiO<sub>2</sub>-patterned vicinal substrates is promising for nanoelectronic device applications such as single-electron memory devices. © 2002 American Institute of Physics. [DOI: 10.1063/1.1534385]

Self-assembled quantum dots (SAQDs) via the Stranski–Krastanow growth mode have been demonstrated to be defect free and to have high density with three-dimensional quantum confined nature of the electronic spectra. Even though they produce these properties for the realization of quantum functional electron devices,<sup>1–3</sup> they are randomly distributed with fluctuations in their size as well as position, when SAQDs are formed on a planar substrate. This randomness is undesirable particularly for electronic device applications. Thus, there has been a considerable effort to manipulate the position, size, and density of SAQDs using various approaches.

Recently, *in situ* fabrication of self-aligned QDs on vicinal (001) GaAs substrate has been studied.<sup>4–7</sup> The average size of QDs perpendicular to the step lines on vicinal substrates was limited by the atomic terrace width, which was given by the period of step bunching formed during the growth of GaAs.<sup>5</sup> The control of the size and interval of InAs SAQDs on vicinal (001) GaAs substrate was successfully manipulated by using these properties.<sup>6,7</sup> However, they have inherent difficulties for the precise control of the size and the position of SAQDs along the step lines and additional fine lithography is still required in order to use SAQDs locally for quantum electronic devices.

On the other hand, several groups have devoted much effort to selective area growth technique using the patterned planar substrates because appropriate patterning of the mask layer and the control of the growth conditions enable us to realize their control without another fabrication damage.<sup>8,9</sup> We have reported the number control of InAs SAQDs on exact (001) GaAs substrates by employing nanometer-scale patterned SiN<sub>x</sub> layers as a mask for the selective growth area.<sup>8</sup> However, the selective growth of SAQDs on patterned exact (001) GaAs substrate have difficulties for the control of

the interval or position of multiple SAQDs, which are also important for quantum electronic device.

These two methods for selective growth of SAQDs have advantages and disadvantages which complement each other as discussed, nevertheless it has not yet been reported about their combination, that is, the selective growth of SAQDs on patterned vicinal substrates. This letter reports the fabrication of single- or double-row aligned In<sub>0.8</sub>Ga<sub>0.2</sub>As SAQDs on the (001) top facet by utilizing SiO<sub>2</sub>-patterned vicinal (001) GaAs substrates. The dependence of the surface morphologies of In<sub>0.8</sub>Ga<sub>0.2</sub>As SAQDs on the width of the (001) top facet [ $W_{(001)}$ ] and the misorientation angles of the substrate were investigated by scanning electron microscopy (SEM) and atomic force microscopy (AFM) measurements.

Starting materials used in this study were 1°, 2°, and 5°-off (001) GaAs with and without patterned SiO<sub>2</sub> as a mask. The direction of misorientation angles is  $[1\bar{1}0]$  and the thickness of SiO<sub>2</sub> is 20 nm. The whole patterns within 1900×1900 μm<sup>2</sup> consisted of 25 kinds of pattern regions. Each pattern was filled with stripe windows (wire region) along  $[1\bar{1}0]$  direction in 800 nm periodicity within 100×100 μm<sup>2</sup>. 25 kinds of patterns have different width of opening region ( $W_0$ ) which were varied from 300 to 700 nm. The selective area was patterned by electron beam lithography and wet chemical etching technique, and SiO<sub>2</sub> outside of the pattern definition was totally removed by photolithography and wet chemical etching.

The growth of GaAs buffer layer and In<sub>0.8</sub>Ga<sub>0.2</sub>As SAQDs were performed by low-pressure metalorganic vapor phase epitaxy (MOVPE) working at 76 Torr. Purified hydrogen (H<sub>2</sub>) was used as a carrier gas. The source materials used were trimethylgallium (TMG) for GaAs buffer layer, trimethylindium (TMI) and triethylgallium (TEG) for SAQDs, and 20% arsine (AsH<sub>3</sub>) in H<sub>2</sub>. The partial pressures of AsH<sub>3</sub> and TMG for GaAs buffer layer were maintained at 2.5×10<sup>-4</sup> and 7.3×10<sup>-6</sup> atm, respectively. The partial pressure of AsH<sub>3</sub>, TEG, and TMI for In<sub>0.8</sub>Ga<sub>0.2</sub>As SAQDs

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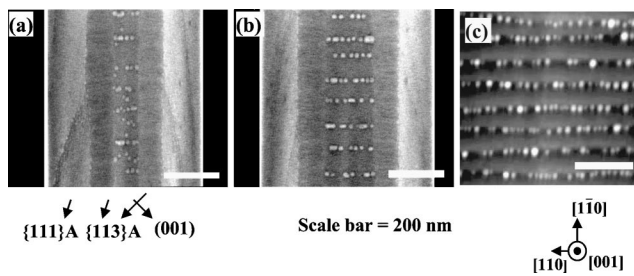


FIG. 1. SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned  $1^\circ$ -off (001) GaAs substrate, when the widths of opening regions ( $W_0$ ) were (a) 560 and (b) 620 nm. (c) AFM image of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on unpatterned  $1^\circ$ -off (001) GaAs substrate.

were  $2.5 \times 10^{-5}$ ,  $5.0 \times 10^{-7}$ , and  $9.1 \times 10^{-7}$  atm, respectively. The growth temperature for the GaAs and  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs were 700 and 500  $^\circ\text{C}$ , and the corresponding nominal thickness were 200 nm and 3.2 ML, respectively.

Figures 1(a) and 1(b) show SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs and GaAs buffer layers grown on  $\text{SiO}_2$ -patterned  $1^\circ$ -off (001) GaAs substrate with  $W_0$  of 560 and 620 nm, respectively. During the growth of the GaAs buffer layer on opening region of  $\text{SiO}_2$ -patterned vicinal (001) GaAs substrates, the formations of GaAs buffer layers have been changed to mesa-structure which consist of (001) top facet, {113} A facets surrounding the the top region, and {111}A facets on side walls. As the growth of GaAs buffer layer proceeds on opening regions, the widths of the (001) top facet [ $W_{(001)}$ ] were directly proportional to  $W_0$  and the corresponding  $W_{(001)}$  were 80 and 153 nm, respectively.  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs were not formed on {111}A and {113}A facet, and grew selectively on (001) top facet. As shown in Figs. 1(a) and 1(b), the formations of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs were changed by  $W_{(001)}$ . The number of SAQDs perpendicular to the  $[1\bar{1}0]$  misorientation direction on (001) top facet was increased by the enhanced  $W_{(001)}$ . The interval of SAQDs along the  $[1\bar{1}0]$  misorientation direction on (001) top facet ( $d_{\parallel}$ ) was not clear in Fig. 1(a), whereas it was clearly distinguished in Fig. 1(b). Also, the average  $d_{\parallel}$  as a function of  $W_{(001)}$  ( $\bar{d}_{\parallel}$ ) in Fig. 1(a) was narrower than in Fig. 1(b). Figure 1(c) shows an AFM image of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on unpatterned  $1^\circ$ -off (001) GaAs substrate with the same growth conditions of GaAs buffer layer and SAQDs. SAQDs were formed on step lines transformed by bunching effect of GaAs buffer layer due to the surface migration length of Ga adatoms along the  $[1\bar{1}0]$  misorientation direction of the substrate ( $L_{\parallel}$ ).<sup>7,10</sup> The interval of step lines was 67 nm and it was in accordance with  $d_{\parallel}$  in Fig. 1(b). Therefore, it can be confirmed that  $L_{\parallel}$  on the (001) top facet in Fig. 1(b) is in accordance with that in Fig. 1(c), whereas  $L_{\parallel}$  on the (001) top facet in Fig. 1(a) is smaller than that in Fig. 1(c). There is no clear explanation about the reason yet, but we believe this may be caused by the dissimilar interfacet migration of Ga adatoms on different facets, when  $W_{(001)}$  is changed.<sup>11</sup>

Figures 2(a), 2(b), and 2(c) show SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs and GaAs buffer layers grown on  $\text{SiO}_2$ -patterned  $2^\circ$ -off (001) GaAs substrate with  $W_0$  of 547 and 560 nm, and an AFM image of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on unpatterned  $2^\circ$ -off (001) GaAs substrate, respec-

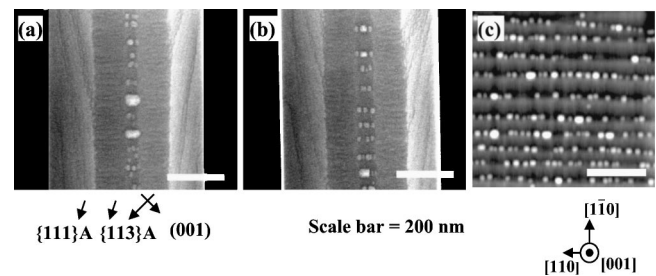


FIG. 2. SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned  $2^\circ$ -off (001) GaAs substrate, when the widths of opening regions ( $W_0$ ) were (a) 547 and (b) 560 nm. (c) AFM image of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on unpatterned  $2^\circ$ -off (001) GaAs substrate.

tively. The corresponding  $W_{(001)}$  in Figs. 2(a) and 2(b) were 43 and 52 nm, respectively.  $\bar{d}_{\parallel}$  in Fig. 2(b) and the interval of step lines in Fig. 2(c) have the same width of 52 nm.  $\bar{d}_{\parallel}$  in Fig. 2(a) was narrower than that in Fig. 2(b). Therefore, the relation between the formation of SAQDs and  $W_{(001)}$  was similar to that on  $\text{SiO}_2$ -patterned  $1^\circ$ -off (001) GaAs substrate. However,  $W_{(001)}$  having definite  $d_{\parallel}$  for  $2^\circ$ -off (001) GaAs substrate [in Fig. 2(b)] was narrower than that for  $1^\circ$ -off GaAs substrate [in Fig. 1(b)].

Figure 3 shows the experimental results for  $d_{\parallel}$  as a function of  $W_{(001)}$ . The filled triangles and open circles represent the experimental results for  $d_{\parallel}$  of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned  $1^\circ$ -off and  $2^\circ$ -off (001) GaAs substrates, respectively. The enhanced  $\bar{d}_{\parallel}$  was caused by the enhanced  $W_{(001)}$ , so that it saturated to the same width as interval of step lines on unpatterned vicinal substrates. We found that large fluctuation in  $\bar{d}_{\parallel}$  value occurred up to a certain value of  $W_{(001)}$  after which  $\bar{d}_{\parallel}$  value saturated. This fluctuation region of  $\bar{d}_{\parallel}$  for  $1^\circ$ -off substrate ( $\sim 150$  nm) (I) is wider than that for  $2^\circ$ -off substrate ( $\sim 50$  nm) (II) as shown in Fig. 3. For quantitative understanding, we fit the experimental results in the following equation:

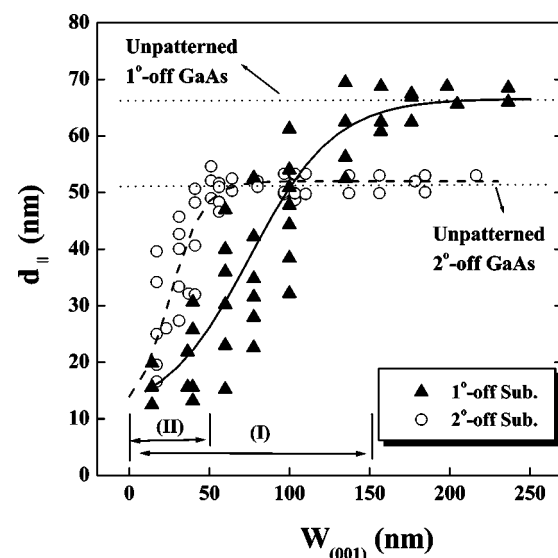


FIG. 3. The experimental results for the interval SAQDs along the misorientation direction of substrate on (001) top facet ( $d_{\parallel}$ ) as a function of the width of (001) top facet ( $W_{(001)}$ ). The solid line and dashed line represent the fitting curves of average  $d_{\parallel}$  ( $\bar{d}_{\parallel}$ ) for  $1^\circ$ -off and  $2^\circ$ -off (001) GaAs substrates, respectively.

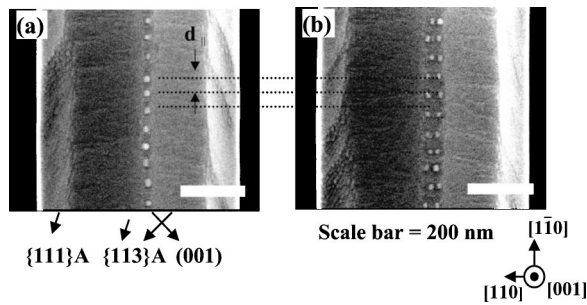


FIG. 4. SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned  $5^\circ$ -off (001) GaAs substrate, when the widths of opening regions ( $W_0$ ) were (a) 628 and (b) 650 nm, respectively.

$$\bar{d}_{\parallel} = \frac{\bar{d}_{\min} - \bar{d}_{\max}}{1 + \exp[(W_{(001)} - \bar{W}_{(001)})/\beta]} + \bar{d}_{\max}, \quad (1)$$

where  $\bar{d}_{\max}$ ,  $\bar{d}_{\min}$ ,  $\bar{W}_{(001)}$ , and  $\beta$  represent maximum  $\bar{d}_{\parallel}$ , minimum  $\bar{d}_{\parallel}$ , the half width of maximum  $W_{(001)}$  in which fluctuation of  $\bar{d}_{\parallel}$  occurs, and a proportional constant decided from the slope of the fitting curves, respectively. The values of  $\bar{d}_{\min}$ ,  $\bar{d}_{\max}$ ,  $\bar{W}_{(001)}$ , and  $\beta$  for the experimental results were 11, 67, 75, and 27 nm for  $1^\circ$ -off (001) GaAs substrate, and 11, 52, 25, and 11 nm for  $2^\circ$ -off, respectively. The solid and dashed lines in Fig. 3 represent the fitting curves for  $1^\circ$ -off and  $2^\circ$ -off (001) GaAs substrates, respectively. According to the experimental results,  $\beta$  decided from the slope of the fitting curves was related to the misorientation angles of the substrates. The value of  $\beta$  and the fluctuation region for  $d_{\parallel}$  were reduced, as the misorientation of substrates was larger. There is no clear explanation yet about the relation between the misorientation angles of the substrate and  $\bar{d}_{\parallel}$ , but we also believe this to be caused by the dissimilar interfacet migration of Ga adatoms on different facets, when the misorientation angles of the substrates are changed.

In order to reduce the fluctuation regions for  $d_{\parallel}$  which are undesirable for electronic device applications, we applied the same growth condition to  $\text{SiO}_2$ -patterned  $5^\circ$ -off (001) GaAs substrate. Figures 4(a) and 4(b) show SEM images of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned  $5^\circ$ -off (001) GaAs substrate with  $W_0$  of 628, and 650 nm, respectively. The corresponding  $W_{(001)}$  were 19 and 46 nm, respectively. Single-row aligned SAQDs in Fig. 4(a) and double-row

aligned SAQDs in Fig. 4(b) have the same  $\bar{d}_{\parallel}$  of 50 nm and the fluctuation region for  $d_{\parallel}$  was not observed. Therefore, single- or double-row aligned SAQDs having definite  $\bar{d}_{\parallel}$  was successfully fabricated by utilizing  $\text{SiO}_2$ -patterned  $5^\circ$ -off (001) GaAs substrate and a reduction of the fluctuation region for large misorientation angle was confirmed.

In summary, the interval of  $\text{In}_{0.8}\text{Ga}_{0.2}\text{As}$  SAQDs grown on  $\text{SiO}_2$ -patterned vicinal GaAs (001) substrates by SA-MOVPE can be controlled by  $W_{(001)}$  and the misorientation angle of the substrate. In order to maintain definite  $d_{\parallel}$  on (001) top facet equal to the interval of step lines on unpatterned vicinal (001) GaAs substrate, it is effective to use vicinal substrate having higher misorientation angles, single- or double-row aligned SAQDs having definite  $\bar{d}_{\parallel}$  were successfully fabricated by utilizing  $5^\circ$ -off (001) GaAs substrate. These results suggest that the selective growth technique of SAQDs by utilizing  $\text{SiO}_2$ -patterned vicinal substrates is promising for nanoelectronic device applications, such as single-electron memory devices.

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