Spatial Point Analysis of Quantum Dot Nucleation Sites on InAs Wetting Layer

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We perform spatial point analysis of InAs quantum dot nucleation sites and surface reconstruction domain pattern on an InAs wetting layer, giving insights for quantum dot nucleation mechanism. An InAs wetting layer grown to 1.5 monolayers in thickness on a GaAs(001) substrate has been observed at 300 °C by using in situ scanning tunneling microscopy. The surface exhibits (1 × 3)/(2 × 3) and (2 × 4) reconstruction domains. A nearest-neighbor analysis finds that point pattern of quantum dot precursors was more similar to that of (1 × 3)/(2 × 3) domains which are specific to Ga-rich region. This provides the evidence that InAs quantum dot nucleation is induced by Ga-rich fluctuation within an InAs wetting layer.

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Quantum dots (QDs) are potentially used in semiconductor laser devices and single photon sources of quantum computation and quantum communication [1–4]. Although it has been pointed out that highly dense and uniform QD arrays are essential for the efficiency of the devices, little is known of the growth mechanism of QDs to control the nucleation sites on a Stranski-Krastanow (SK) grown wetting layer (WL). Some atomic-level theoretical studies on dynamics of surface atoms have been carried out to understand the growth mechanisms [5–8]. First principle calculations showed that the migration barrier energy of In adatom on GaAs(001) surface is higher than that on 1ML-InAs/GaAs(001) [6, 7]. Using kinetic Monte Carlo (kMC) simulations [8], Tsukamoto et al. found that some migrating In adatoms were captured on Ga-rich fluctuation, within an In/Ga mixed layer, to become a nucleation site [9]. To the best of our knowledge, however, there has not been reported any direct evidence that alloy fluctuation becomes a QD nucleation site. It is still vital to investigate WL surface in an atomic scale, in particular the surface reconstruction, preceding QD nucleation.

Since surface reconstruction changes microscopically and dynamically in the course of WL growth [10–12], in situ scanning tunneling microscopy (STM) during molecular beam epitaxy (MBE) growth at high temperatures, such as STMBE [13], is one of powerful tools to observe it. It is reported that fast Fourier transform analysis of atomic-scale in situ STM images of InAs WL on a GaAs(001) substrate, as well as reflectance high-energy electron diffraction (RHEED) measurements, has revealed that the surface reconstruction changes from c(4 × 4) to the mixed structure of (1 × 3)/(2 × 3) domains and (2 × 4) domains prior to QD formation [9].

Structure models of (1 × 3)/(2 × 3) and (2 × 4) surface reconstructions have been investigated by many researchers using core-level photoemission spectroscopy [14], reflectance-difference spectroscopy [15], ab initio calculations in a local density approximation [6].

FIG. 1. (Color online) Schematic diagrams of surface reconstruction models of InAs/GaAs wetting layer on GaAs(001) reproduced from literature: (a) (1 × 3) [14], (b) (1 × 3) [15], (c) (2 × 3) [9], (d) α2(2 × 4) [16, 17], (e) β2(2 × 4) [16, 17]. Background shade indicates unit cell.

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that is a regular (ordered) pattern, a Poisson (random) pattern, and a clustered (aggregated) pattern [18, 19]. In a regular pattern, points are distributed uniformly. A Voronoi tessellation, that is polygonal decomposition of a space by perpendicular bisector lines among neighboring points, is often used in spatial point analysis. The standard deviation of Voronoi cell area, \( \sigma_{Vc} \), represents point patterns. Here, we have to be careful of the Voronoi cells touching the boundary of the study region because of “edge effect” that contribution from points outside cannot be taken into account. In this study, such invalid Voronoi cells were excluded from the study region. For more precise analysis, second-order properties of point patterns like nearest-neighbor distances are useful [19–22]. Let \( r \) denote the distance to the nearest point from a randomly selected location in the study region \( R \). The \( F(d) \) function denotes the cumulative frequency distribution of \( r \) [19] and hence the probability that \( r \) occurs less than any particular distance \( d \). Since the \( F \) function is practically identical to the probability of plotting a random point within any of circles \( C_i(d) \), of each radius \( d \), centered on each of the points, it is simply computed by

\[
F(d) = \frac{\text{Area } (\bigcup C_i(d) \cap R)}{\text{Area } (R)}, \tag{1}
\]

where the numerator is the area of the union of the circles, and the denominator is the area of the study region \( R \). To compare \( F(d) \) between different study regions, \( d \) should be normalized by the factor \( f \) as follows:

\[
f = \sqrt{\frac{\text{Area } (R)}{N}}, \tag{2}
\]

where \( N \) is the number of points [20].

A piece \((11 \times 13 \times 0.6 \text{ mm}^3)\) of GaAs(001) crystal was used as a substrate. First the surface was thermally cleaned to remove the oxide layer under \( 1 \times 10^{-4} \text{ Pa} \) of an \( \text{As}_4 \) atmosphere in an MBE growth chamber. Next, a GaAs buffer layer was grown on the surface by using MBE until atomically smooth surface was obtained. The substrate was annealed at 430 \(^\circ\text{C}\) for 0.5 h to confirm the formation of \( c(4 \times 4) \) reconstruction with RHEED. An STM unit was transferred to the sample holder in the growth chamber and started scanning. This means that the sample was neither cooled to room temperature nor exposed to air to be observed with STM. The tip bias was -3 V and the tunnel current was 0.2 nA. A flux of In was irradiated at the InAs growth rate of \( 2.5 \times 10^{-4} \text{ MLs}^{-1} \). After 1.5 monolayer (ML) of InAs WL growth, the substrate temperature was decreased to 300 \(^\circ\text{C}\) and the \( \text{As}_4 \) flux was shut off so that the surface reconstruction should be observed in an atomic scale. Another samples was prepared in the same way but SK growth of InAs WL was continued at 430 \(^\circ\text{C}\) until QD precursors were formed [3]. An STM image of QD precursors was recorded to analyze the distribution of QD nucleation sites.

Figure 2 shows the filled-state STM image of 1.5 ML of InAs WL recorded at 300 \(^\circ\text{C}\). The image seems more or less scratched because of migrating In adatoms on the surface but shows the stripes due to As dimers clearly enough to measure the pitch. The stripe pitch corresponds to the unit cell length along [110] azimuth of InAs WL surface reconstructions in Fig. 1. Although it is difficult to discuss the unit cell size of strained and mixed surface reconstructions, it is expected to have some intermediate value between those of GaAs and InAs. We assumed that the stripe pitch along [110] azimuth ranged 0.6–1.0 nm for \( c(4 \times 4) \), 1.0–1.4 nm for \((1 \times 3)/(2 \times 3)\), and 1.4–2.0 nm for \((2 \times 4)\). Before measuring the stripe pitch, the STM image [Fig. 2] was divided by a \( 25 \times 25 \) mesh. As shown in the schematic diagram in Fig. 3 the size of each mesh cell is 1.2 nm which is comparable to the unit cell sizes of the InAs WL surface reconstructions. The stripe pitch was measured from the STM line...
FIG. 4. (Color) Surface reconstruction domains, indicated by oval markers, and Voronoi tessellations of (a) \((1 \times 3)/(2 \times 3)\) and (b) \((2 \times 4)\) surface reconstruction maps.

profile along [110] azimuth for each mesh cell. The data are plotted in the color map of Fig. 3. Most cells show \((1 \times 3)/(2 \times 3)\) or \((2 \times 4)\) surface reconstruction although some cells are blank because of step edges.

It is predicted that Ga-rich fluctuation, comprised of at least eight Ga atoms in an In/Ga mixing layer, should be a QD nucleation site according to kMC simulations [9]. For such Ga-rich fluctuation to be formed, four unit cells of \((1 \times 3)/(2 \times 3)\) need to be contiguous, each of which has one or two Ga atoms. In this study, groups of four neighboring mesh cells, having the same surface reconstruction, were located in the map [Fig. 3] and indicated by oval markers in Fig. 4. For each of \((1 \times 3)/(2 \times 3)\) and \((2 \times 4)\) surface reconstructions, the centroid points of the domains were marked and their coordinates were measured by using ImageJ software [23, 24]. The centroid coordinates were used for the Voronoi tessellations of the reconstruction domain maps [Fig. 4] and computation of the \(F\) function [20].

Figure 5 shows the 150 nm × 150 nm STM image of InAs QD precursors immediately after nucleation [9] and Voronoi tessellations computed for each of 3 × 3 regions.

![FIG. 5. (Color) 150 nm × 150 nm STM image of QD precursors nucleated on SK grown InAs WL at 430°C and Voronoi tessellations computed for each of 3 × 3 regions.](image)

| Domain Type                  | \(\rho\) \((10^{12} \text{ cm}^{-2})\) | \(\sigma_{Vc}\) |
|-----------------------------|--------------------------------------|-----------------|
| \((1 \times 3)/(2 \times 3)\) | 6.2                                  | 0.38            |
| \((2 \times 4)\)            | 4.2                                  | 0.31            |
| QD precursors               | 0.96–1.7                             | 0.20–0.59       |

Both traces of \((1 \times 3)/(2 \times 3)\) and \((2 \times 4)\) domains were similar to the envelope of QD precursors although they differ in the detail. The traces of the surface reconstruction domains and the QD precursors were plotted between those of the ordered pattern and the Poisson patterns. This shows that surface reconstruction domains and QD precursors are distributed in a rather ordered pattern than a random pattern. An ordered pattern likely occurs when there is repulsive force among points. It is difficult to discuss the origin of repulsive force by STM images, one can consider the surface strain distribution. First principles calculations of WL would give some insights for the surface strain distribution. It is possible that the uniformity of QD nucleation, which is rather ordered, is originated from the distribution of surface reconstruction domains.

The trace of \((2 \times 4)\) domains is located rather ordered in the QD envelope, and completely deviates from the QD envelope in small \(d\) region as can be seen in the magnified view in Fig. [9] The trend of \(F(d)\) where \(d\) is small...
is dependent on the pattern in dense-point areas. On the other hand, the trend of $F(d)$ where $d$ is large is dependent on the pattern in sparse-point areas. The pattern in dense-point areas is particularly crucial because of high surface strain which severely affects the distribution. In the sense of not deviating in dense-point areas, the trace of $(1 \times 3)/(2 \times 3)$ domains represents the QD precursor envelope better than that of $(2 \times 4)$ domains. In other words, the distribution of QD precursors are more similar to that of $(1 \times 3)/(2 \times 3)$ domains than that of $(2 \times 4)$ domains. The similarity in the density and the point pattern implies that QD nucleation is related to $(1 \times 3)/(2 \times 3)$ domains where Ga-rich fluctuation is assumed to be formed. This also means that QD nucleation sites are already determined at the time when InAs WL is grown to 1.5 ML or before.

In conclusion, we have shown, by using in situ STM, the similarity in the density and the spatial point patterns between $(1 \times 3)/(2 \times 3)$ surface reconstruction domains on 1.5 ML of SK grown InAs WL and QD precursors nucleated. This implies that QD nucleation site is related to the distribution of $(1 \times 3)/(2 \times 3)$ domains at the stage of 1.5 ML-grown InAs WL. Since a $(1 \times 3)/(2 \times 3)$ domain composed of four unit cells is assumed to contain Ga-rich fluctuation with at least eight Ga atoms, this model provides a consistent evidence that QD nucleation is induced by tiny alloy fluctuation as predicted by kMC simulations [9]. The mechanism of QD nucleation, exhibited here, has important technological implications for the self-assembly and the artificial arrangement of QDs.

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