Effects of a Si Molecular Beam on the Formation of InAs Quantum Dots

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We have investigated the effect of a Si molecular beam on self-assembled InAs quantum dots (QDs) on GaAs grown by molecular beam epitaxy. We changed the Si coverage (Θ) and the substrate temperature and monitored their effects on the InAs QDs by using atomic force microscopy. The 2D-3D transition of self-assembled InAs QDs was found to be delayed by the existence of Si-adatoms. As the Si coverage (Θ) increased, the density of QDs increased, but after a critical Si coverage (Θ=0.12), the density decreased rapidly due to the coalescence of dots. This indicates that Si atoms act as nucleation centers for the formation of self-assembled InAs QDs. By optimizing the Si coverage (Θ) and the substrate temperature, we could obtain InAs QDs with high density.

I. INTRODUCTION

Self-assembled InAs quantum dots (QDs) have been the subject of much research due to their potential in electronic and optoelectronic device applications [1]. The formation of InAs QDs on GaAs has the advantage that the growth rate can be precisely controlled; also, it is relatively easy. However, self-assembled InAs QDs also have several problems, such as low density, nonuniformity, and the difficulty of size and position control. Thus, there have been many endeavors to overcome the problems: modification of growth conditions, such as substrate temperature [2], growth rate [3], V/III ratio [3,4] and substrate misorientation [5,6], selective area growth [7], and the use of a surfactant [8]. In the case of a surfactant, its effect led to modifications of the growth mode and the size distribution of QDs. For instance, it was reported [8] that, if Te was used as a surfactant, the growth mode transition and relaxed island formation occurred in the growth of self-assembled InAs QDs. Also, narrow size distributions of QDs were obtained through the selection of a suitable InAs thickness. However, when Te was used, the PL intensity decreased due to the effect of band filling [9].

We selected a Si molecular beam to use Si as a surfactant and expected that the Si molecular beam would modify the morphology of InAs QDs. The InAs thickness, the Si coverage (Θ), and the substrate temperature during the Si irradiation were varied, and their effects were monitored by using atomic force microscopy (AFM). We could obtain high density of InAs dots by optimizing the growth parameters. Also, by measuring the photoluminescence (PL), we investigated the effects of the Si molecular beam on the optical properties of self-assembled InAs QDs.

II. EXPERIMENTAL PROCEDURE

All samples were grown on semi-insulating GaAs (100) substrates by using the molecular beam epitaxy technique. The growth sequence was; a 0.5-µm GaAs buffer layer, Si-molecular beam irradiation, and an InAs QD layer. The substrate temperature was 590 °C and 490 °C during the buffer layer growth and the QD layer growth, respectively, and two different substrate temperatures of 590 °C and 490 °C were employed during the Si beam irradiation. The temperature of the Si cell was maintained at 1120 °C. The InAs thickness was varied from 2 to 3 monolayers (MLs) by controlling the shutter of the In cell, and the Si coverage (Θ) was controlled by the irradiation time.

The 2D to 3D growth mode transition was investigated by observing the reflection high-energy electron diffraction (RHEED) pattern, and the surface morphology was observed by using AFM measurements. The PL measurement was performed using an Ar-ion laser and a liquid-nitrogen-cooled Ge detector. We performed the PL measurement with samples which had different Si coverages (Θ=0, 0.004, and 0.013). Also, using two
Fig. 1. 0.5 × 0.5 μm² AFM images of samples with various InAs thicknesses: (a) 2 MLs, (b) 2.3 MLs, (c) 2.5 MLs, and (d) 3 MLs. The substrate temperature was 590 °C during the irradiation with the Si molecular beam.

For samples, Si coverages (Θ) of 0 and 0.004, we performed temperature-dependent PL experiments.

III. RESULTS AND DISCUSSION

We changed the InAs thickness from 2 to 3 MLs after irradiation with a Si beam corresponding to a Si coverage (Θ) of 0.078. Figure 1 shows 0.5 × 0.5 μm² AFM images of samples with various InAs thicknesses. In Fig. 1(a) with a 2-ML InAs thickness, QDs are rarely observed. This indicates that the wetting layer in the growth of self-assembled InAs QDs using a Si molecular beam must be thicker than 2 MLs. QDs begin to appear gradually at a 2.3-ML InAs thickness and tend to be mature at 2.5 MLs, as shown in Figs. 1(b) and 1(c). The densities of the QDs are 2.3 × 10¹⁰ cm⁻² and 5.4 × 10¹⁰ cm⁻², respectively. For a 3-ML InAs thickness (Fig. 1(d)), incoherent islands are formed by aggregation of the QDs. In other words, growth beyond 2.5 MLs leads to the formation of large islands with a drastic reduction in density.

It has been reported [10–12] that, without Si-beam irradiation, self-assembled InAs QDs are formed near a 1.7-ML InAs thickness, which is called the critical thickness. Because the critical thickness is 2.3 MLs in our experiments, we conclude that the 2D-3D transition was delayed by the Si-beam irradiation. The Si molecular beam is considered to act as a surfactant which promotes the 2D growth mode. The growth mode can be interpreted by the site-exchange mechanism of Si and As atoms [13–15]. The Si-covered surface adsorbs As atoms and exchange occurs between the surface As layer and the Si sublayer. Si atoms prevent the deposition of As atoms and reduce the surface free energy of As atoms, so finally the 3D growth of InAs is suppressed.

Next, we grew six samples (A-F) with different Si coverages (Θ) of 0, 0.026, 0.039, 0.078, 0.117, and 0.156. The substrate temperatures was fixed at 590 °C during Si-beam irradiation, and the InAs thickness was 2.5 MLs for all samples. Table 1 presents the AFM statistical data, average lateral size, average height, and density of QDs, for the six samples. Among the data, the average lateral size and the density of QDs are plotted in Fig. 2 as a function of the Si coverage (Θ). The density of QDs tends to increase as the Si coverage (Θ) increases in the range up to Θ=0.117, but it decreases rapidly for Θ>0.117. On the other hand, the lateral size of the

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Si coverage (Θ)</th>
<th>average lateral size (nm)</th>
<th>Average height (nm)</th>
<th>Aspect ratio (height/lateral size)</th>
<th>Integrated volume (nm³)</th>
<th>Density (cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0</td>
<td>36.69</td>
<td>3.59</td>
<td>0.090</td>
<td>4.0 × 10⁴⁴</td>
<td>1.8 × 10¹⁰</td>
</tr>
<tr>
<td>B</td>
<td>0.026</td>
<td>30.09</td>
<td>2.40</td>
<td>0.080</td>
<td>3.6 × 10⁴⁴</td>
<td>4.2 × 10¹⁰</td>
</tr>
<tr>
<td>C</td>
<td>0.039</td>
<td>27.77</td>
<td>1.57</td>
<td>0.057</td>
<td>2.2 × 10⁴⁴</td>
<td>4.7 × 10¹⁰</td>
</tr>
<tr>
<td>D</td>
<td>0.078</td>
<td>27.34</td>
<td>1.76</td>
<td>0.064</td>
<td>2.8 × 10⁴⁴</td>
<td>5.4 × 10¹⁰</td>
</tr>
<tr>
<td>E</td>
<td>0.117</td>
<td>19.17</td>
<td>0.79</td>
<td>0.041</td>
<td>8.2 × 10⁴³</td>
<td>7.2 × 10¹⁰</td>
</tr>
<tr>
<td>F</td>
<td>0.156</td>
<td>38.14</td>
<td>6.46</td>
<td>0.169</td>
<td>2.8 × 10⁴⁴</td>
<td>7.4 × 10⁹</td>
</tr>
</tbody>
</table>
QDs decreases up to $\Theta=0.117$ and increases rapidly for $\Theta>0.117$.

At zero Si coverage ($\Theta$), the dots begin to form at a 1.7-ML InAs thickness, and at a thickness of 2.5 MLs, larger islands are already formed with the relaxation of small QDs [10]. As described earlier, with Si irradiation, QD formation occurs at a higher InAs thickness. We suggest that the Si atoms act as nucleation centers; InAs forms new QDs at these nucleation centers rather than enlarging the existing QDs. Therefore, as the Si coverage ($\Theta$) increases, the density of QDs increases while the lateral size and the height decrease. However, a critical Si coverage ($\Theta$) exists, above which the QDs become larger by coalescence among QDs (sample F). Thus, the critical Si coverage ($\Theta=0.12$) is the optimum condition for the formation of a high density of QDs.

It is interesting to note that the number of nucleation centers is not directly proportional to the amount of Si coverages ($\Theta$). This can be explained by an analysis of the aspect ratios (ratio of height relative to lateral size) of the QDs and the volumes of QDs indicated in Table 1. As Si coverage ($\Theta$) increases, not only does the density of QDs increase, but also the aspect ratio becomes smaller. Considering the slopes in the region of $\Theta=0.026$ – 0.117 (samples B-E), excluding the region of $\Theta=0$ – 0.026, the slope is steeper in the region of rapidly decreasing aspect ratio [Fig. 2]. In other words, samples B, C and D, which have relatively low densities compared with sample E, have large aspect ratios (height is high). This means that QDs are overlaid at limited nucleation centers in these samples. The trend of QD size and density in Fig. 2, which is not linear, can also be understood by these aspect ratios. Comparing the QD volumes of five samples (samples A-E), we were able to anticipate that increased Si atoms prevented deposition of As atoms and that As and In atoms irradiating from the knudsen cell were evaporated near the substrate. This is consistent with the result of Fig. 1. Because the QD volume lessens and the nucleation centers increase, the height of QDs comes to be relatively low. From this result, we can say that, to some extent smaller QDs are formed as the Si coverage ($\Theta$) increases. However, critical InAs thickness exists, above which the QDs become larger by coalescence among QDs (sample F). As a result, we have shown that the density and the size of self-assembled QDs can be controlled to some extent by irradiation with a Si molecular beam.

We lowered the substrate temperature during Si irradiation from 590 $^\circ$C to 490 $^\circ$C. Figure 3 shows a 1×1 $\mu$m$^2$ AFM image of(80,180),(410,470) the sample at a Si coverage ($\Theta$) of 0.078. As can be shown in this figure, the density of QDs is about 7.6×10$^{10}$ cm$^{-2}$, which is higher than 5.4×10$^{10}$ cm$^{-2}$ in the case of 590 $^\circ$C. Figure 4 compares the QD size distributions between the two temperatures. The size is smaller and more uniform in the case of 490 $^\circ$C. Thus, we have shown that the substrate temperature during Si-beam irradiation is also an important parameter for obtaining InAs QDs with high density and better uniformity.

Next, we present the photoluminescence (PL) results. Figure 5 shows the PL signal of three samples with different Si coverages ($\Theta$). The PL signal for the sample with $\Theta=0.004$ is much weaker than the signal of the as-grown sample, and a very weak PL signal was observed for $\Theta=0.013$. This fact indicates that Si affects the for-
The photoluminescence spectra, taken at 16K, of samples for Si coverages ($\Theta$) of 0, 0.004, and 0.013.

The activation energies from the fitted results are 51.7 and 24.6 meV, respectively, for $\Theta=0$ and 0.004. Generally, it is said that the larger dot size has the larger activation energy [16]. Therefore, we can say that the sample for $\Theta=0.004$ has smaller dots than that for $\Theta=0$. This fact is consistent with the result of Fig. 2.

IV. CONCLUSIONS

We have investigated the effect of a Si molecular beam on the growth of self-assembled InAs quantum dots. The Si molecular beam delays the formation of QDs by suppressing the 3D growth. As the Si coverage ($\Theta$) increases, the density of QDs increases while their size decreases. This can be explained by assuming that Si atoms act as nucleation centers for the formation of self-assembled InAs QDs. However, after a critical Si coverage ($\Theta$) of 0.12, the density decreases rapidly due to the coalescence of QDs. The density of QDs is also sensitive to the substrate temperature during Si molecular-beam irradiation. This means that the density and the size of self-assembled QDs can be controlled reproducibly by changing the Si coverage ($\Theta$) and the substrate temperature during Si irradiation. Also, the PL intensity decreases as the Si coverage ($\Theta$) increases, and the activation energy of the Si-irradiated sample is smaller than that of the as-grown sample.

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REFERENCES


