Polarization Symmetry of Vertical Photoluminescence from Columnar InAs/GaAs Quantum Dots

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We report the polarization symmetry of vertical photoluminescence from columnar InAs/GaAs quantum dots (QDs) that were formed by stacking small InAs islands directly in the growth direction. QD samples were grown with various stacking numbers using molecular beam epitaxy. We investigated the polarization dependence of the vertical photoluminescence intensity. The polarization dependence was enhanced by increasing the stacking number. We also found that the polarization direction shifted at random from the [110] direction both with the stacking number and the measurement position on the sample. Transmission electron microscopy observations suggested that the polarization features might be governed by problems in the growth process such as the bending of the stacking direction during the formation of QDs with a high aspect ratio.

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I. INTRODUCTION

Self-assembled quantum dots (QD) have been actively studied with a view to their use in optical telecommunication systems and quantum information processing. In particular, QD symmetry has recently attracted research interest since it strongly affects the polarization characteristics of optical devices. Once the QD laser had been developed [1–3], the QD semiconductor optical amplifier (QD SOA) became an interesting research target [4, 5]. The QD SOA can realize the simultaneous optical amplification of multi-wavelength signals at high speed. The polarization dependence of the amplification will be eliminated if a symmetric QD structure is available in the transverse direction [6, 7]. The in-plane symmetry of QDs is also important as regards a QD photon emitter launching entangled photon pairs vertically into a free air space [8, 9]. The symmetry of a QD governs the generation processes of two polarized photons, which must be indistinguishable [10, 11]. The well-known Stranski-Krastanow (SK)-type QD is domical or pyramidal and elongated in the [110] direction on a (001) substrate [12–15]. Therefore, a SK-type QD is not symmetric in the horizontal direction in addition to the vertical direction. Due to the asymmetric QD shape, a SK-type QD has not yet produced entangled photon pairs [16].

A method for controlling the carrier confinement strength in the growth direction has been developed by stacking small QDs to form a columnar shape [17]. Columnar QDs are capable of laser oscillation with a low threshold current [18] and can control the polarization of a horizontal optical beam [19]. We have reported a computational study showing that the in-plane asymmetry of the columnar QD structure is improved in wavefunctions of confined carriers by enhancing the overflow of wavefunctions into a stacked multiple wetting layer [20]. Since the overflow is larger in the direction where the quantum confinement is stronger, it offsets the anisotropy of the QD structure.

In this work, we studied the polarization symmetry of vertical photoluminescence (PL) from columnar QDs. The QDs were grown with various stacking numbers using molecular beam epitaxy. We investigated the dependence of the polarization intensity and the polarization direction of the PL on the stacking number.

II. EXPERIMENTS

The samples were grown on (001)-GaAs substrates at 510°C by using solid source molecular beam epitaxy. After 1.8-monolayer (ML) InAs was supplied to form SK-type seed QDs, a set consisting of [0.7-ML InAs]/[3-ML GaAs] was supplied repeatedly to form columnar QDs. During the supply cycles, small InAs islands were directly stacked on the seed QDs as shown in Fig. 1. We prepared four samples and varied the stacking number from 9 to 32.

We measured the PL spectra perpendicular to the sample surface at room temperature using a 633-nm He-Ne laser as a pump. We also employed an InGaAs photomultiplier detector cooled to −80°C. The polarization dependence of the PL was investigated by placing a linear polarizer in front of a 20-cm monochromator. We compared the PL spectra obtained in the most intense polarization direction with those obtained in the 90°-rotated direction.

A depolarizer was integrated into the system behind the polarizer to eliminate the polarization dependence of the measurement system. We expected the direction with the most intense polarization to agree with the direction of the major axis of the QD structure, i.e., the [110] direction. Therefore, we performed measurements of the angle of polarization from the [110] direction. Transmission electron microscopy was used to observe the sample cross-section.
III. RESULTS AND DISCUSSION

Figure 2 shows the PL spectra of the samples. The angle of polarization from the [1¯10] direction is attached to each spectrum. The spectra changed according to the stacking number, \( n \). The main peak is the emission from the ground state, and the peak immediately to the left of the main peak is the emission from the first excited state. We see that the emission peak wavelength lengthens as the stacking number increases. The emission intensity decreased with increases in the stacking number. The PL spectra of every sample were clearly dependent on the polarization direction. The dependence of the ground state emission intensity was considerable, and was comparatively small for the first excited state. As for the emission wavelength, the difference due to the polarization direction was not obvious for the ground state. For the first excited state, the weaker emission peak appeared to have a slightly shorter wavelength.

The polarization dependence of the PL intensity and the polarization angle from the [1¯10] direction provide information about the QD structure. Figure 3(a) shows the relationship between the stacking number and the ratio of the PL intensities of the ground state for two directions, \( I_{90^\circ}/I_{\max} \). We found that \( I_{90^\circ}/I_{\max} \) decreased monotonically as the stacking number increased. Figure 3(b) shows the relationship between the stacking number and the angle of the polarization direction to [1¯10]. The angle has either been increased and decreased, and it does not have simple relationship with the stacking number.

The anisotropy of the QDs appeared clearly in the PL intensity. The polarization characteristics reflect the form of a wavefunction, and this proves that the in-plane form of the ground state in a columnar QD was not symmetrical in all the samples. As regards the perpendicular form, it has been reported that the polarization anisotropy in the transverse direction is eliminated when the number of stacked columnar QDs, \( n \), is increased to 9 [6]. When \( n \) exceeds 9, the superior polarization direction is reversed, and the reversal is enhanced as \( n \) is increased. Therefore, an \( n = 9 \) sample is expected to have the most isotropic form in the transverse dimension. However, the polarization intensity ratio of the vertical emission for \( n = 9 \) was
0.68 as shown in Fig. 3(a). Since the value is almost the same as that of a SK-type QD [21], we assume that the anisotropy was caused by the seed SK-type QD.

As regards the first excited state, the difference in PL intensity was very small, but the anisotropy clearly appeared. In Fig. 2, the emission wavelength of the first excited state in the 90°-rotated direction is comparatively short. This result suggests that the degeneracy of the first excited state was solved by the anisotropy of the QD structure. The behavior of the emission wavelength will be explained by the rather complicated wavefunction distribution in the excited states, but it is not well understood at present. The luminescence intensity suggests that the energy split did not greatly affect the carrier lifetime in the first excited states.

To evaluate the measured results, we computed the influence of the anisotropy of the columnar QD structure on the electron wavefunction symmetry. We know that the overflow of a wavefunction from a QD offsets the anisotropy of the QD structure [20, 21]. The multiple wetting layer in the columnar QD will promote the overflow. The wavefunction was computed with a very simple model using the three-dimensional finite element method based on the analytical continuum approach [12, 22, 23]. We assumed that the QD had a geometrically simple columnar shape. The QD had multiple wetting layers with multiple GaAs interval layers, and we modeled the multiple layers as a uniform single wetting layer with an averaged composition. The thickness of the wetting layer was assumed to be the same as the height of a columnar QD. \( L_{\text{major}} \) and \( L_{\text{minor}} \) are defined as the QD size along the major and minor axes, respectively. \( L_{\text{major}} \) was assumed to be 15 nm [18]. To evaluate the influence of QD height on the calculated results, we assumed a height of 9.5 to 34.0 nm. The material parameters at 300 K were used. Initially, we computed the strain distribution in the structure in three dimensions so that the strain energy was at its minimum value. Then, after calculating the strain-induced modification of the band gap and the effective mass, we solved the Schrödinger equation to obtain the wavefunction in the QD ground state.

We estimated the dependence of the wavefunction symmetry on the stacking number. The calculation suggested that the structural symmetry of 60 % \( (L_{\text{minor}}/L_{\text{major}} = 0.6) \) is improved to 67 % in the wavefunction. This is caused by the counter overflow of the wavefunction to the outside of the asymmetric QD. When \( L_{\text{minor}}/L_{\text{major}} = 0.3 \), the symmetry is improved to 48 %.

The calculation showed that when the stacking number increases the quantum confinement effect decreases and the amount of wavefunction overflow is reduced. As a result, \( L_{\text{minor}}^\phi/L_{\text{major}}^\phi \) becomes small in accordance with the increase in the stacking number. However, the change is one order smaller than the change in the PL anisotropy. We concluded that the simple columnar QD model could not explain the measurements in Fig. 3(a).

We then investigated the position dependence of the polarization characteristics within a wafer. The PL intensity ratio and angle from [110] of the polarization direction at certain measurement positions in an \( n = 23 \) sample are shown in Figs. 4(a) and (b). Figure 4(a) shows that the variation is not small even if the wafer end is disregarded (position 5). Figure 4(b) shows that the angle from [110] is not constant and is unrelated to the position in the wafer.

The calculations and the experiments suggest that the polarization characteristics are dominated by actual QD formation problems such as the non-uniform growth surface conditions. It should be noted that the anisotropy of the PL intensity was clearly enhanced by increasing the stacking number (Fig. 3(a)). One possible explanation is that the polarization was influenced by the accuracy of the perpendicular stacking. Figure 5 shows a cross-sectional image of columnar QDs grown with \( n = 32 \) observed using transmission electron microscopy. We can see that the QDs do not necessarily have a columnar form, and they are not necessarily straightly perpendicular. It is known that, when multilayer QDs are grown using thin interval layers, the correlation of a perpendicular position is not easily maintained with large stacking numbers. The same phenomenon probably occurs in columnar QDs. The perpendicular position gap will degrade the in-plane symmetry of a columnar QD. The randomness with a polarization angle from the [110] direction in Fig. 4(b) supports this explanation since the perpendicular position numbers of QDs during stacking will vary in a wafer.

![Graph and Diagram](attachment:graph.png)
FIG. 5: Cross-sectional image of columnar QDs grown with \( n = 32 \) observed using transmission electron microscopy.

IV. CONCLUSIONS

We investigated the polarization symmetry of vertical PL from columnar InAs/GaAs QDs. The QDs were obtained by stacking small InAs islands directly in the growth direction. We found that the PL intensities from the ground state differed greatly depending on the polarization direction, and that the difference became large as the stacking number was increased. We also found that the polarization direction shifted from [110] with changes in stacking number, and that the shift also varied in a wafer. The observed polarization dependence was so great that it was not explained by a theoretical calculation of an electron wavefunction with a simple columnar QD model. Transmission electron microscopy observation suggests that the polarization characteristics may be related to perpendicular stacking problems that occur during QD growth such as bending of the stacking direction.

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