Extended Abstracts of the 1994 International Conference on Solid State Devices and Materials, Yokohama, 1994, pp. 175-177 **PB-1-10**

Fabrication of GaAs/AlGaAs Quantum Dots by Metal-Organic Vapor Phase Epitaxy on Patterned GaAs Substrates

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We report on a growth process on patterned GaAs (001) substrate during metal-organic vapor phase epitaxy (MOVPE) and a novel approach for the fabrication of AlGaAs/GaAs quantum dot (QD) structures. The patterned substrate have an array of holes on the surface and those holes are partially filled with GaAs by MOVPE growth, followed by GaAs/AlGaAs quantum well structures. Detailed investigation on growth process on such patterned substrates revealed the presence of complicated two-dimensional migration of Ga and Al between different facets. Formation of GaAs dots was directly confirmed by spatially resolved cathodoluminescence measurements.

1 Introduction

The confinement of charged carries into reduced dimensions opens up a possibility to realize high-performance devices and novel physics. For the realization of quantum wires (QWRs) or quantum dots (QDs), an approach utilizing the dynamic self-organizing mechanisms present in the crystal growth is more promising, as compared with the direct lithography-and-etching approach, since lower defect densities and automatic size reduction by atom migration can be expected.

We report here on a novel approach toward realization of QDs by metal-organic vapor phase epitaxy (MOVPE) growth. The essence of the present fabrication method is to use patterned GaAs (001) substrates on which an array of holes are defined, and to fill them up with AlGaAs/GaAs layers by MOVPE growth. Three dimensional quantum confinement can realized by the combination of lateral confinement scheme reported in Ref. [1] or [2] in the two orthogonal directions and vertical confinement of conventional quantum wells. As compared with the QD arrays first reported by Fukui *et al.*[3], the present approach offers an important advantage that it allows easier introduction of coupling between QDs (coupled QDs) and adjacent quantum wells (QWs), which is important to realize smooth transfer of electrons or efficient carrier injection into QDs.

In this study, we first investigated the MOVPE growth process on patterned substrates to discuss on the feasibility of the present approach. Then, to confirm the formation of dot structures, spatially resolved cathodoluminescence (CL) measurements were carried out and their results will be described.

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2 Experimental Procedure

GaAs (001) patterned substrates having an array of bath-tub-like holes as schematically shown in Fig. 1(a) were prepared by standard photolithography and wet chemical etching. Here, the period of the array is 4μm. Though the initial openings of photoresist mask is square, the holes after etching becomes rectangular because of the crystallographic anisotropy. This also results in the appearance of forward mesa structures consisting of two (111)A facets in the [110] (Y–Y′ of Fig. 1(a)) direction, and both forward and reversed mesa structures in the [1$\bar{1}$0] (X–X′ of Fig. 1(a)) direction, as evident in a secondary electron microscope (SEM) image of Fig. 1(b).

MOVPE growth were carried out using horizontal reactor at a pressure of 0.1atm with source material of trimethylgallium (TMGa), triethylaluminum (TEAl), and AsH_3. The growth temperature was 750°C. The growth dynamics on patterned substrates were investigated by cross sectional SEM observation of AlGaAs/GaAs layered structures, which consist of 10nm-thick AlGaAs and 90nm-thick GaAs with total thickness of 1μm. For the characterization of optical properties, QWs with thickness of 8nm were grown after a 1.5μm-thick buffer layer and cathodoluminescence (CL) measurements were carried out at 11K with acceleration voltage of 5keV and beam current of 50pA.

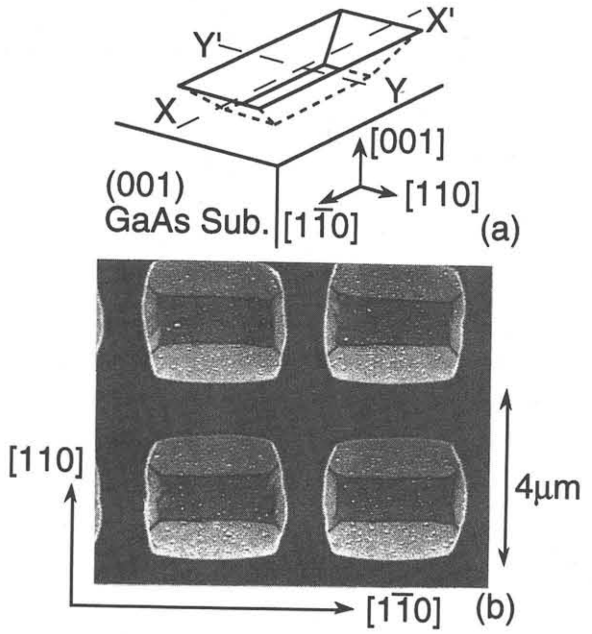

Fig. 1: (a) Schematic representation of a hole used for growth in the present study. (b) Plane-view secondary electron microscope (SEM) image of the patterned substrate

3 Results and Discussions

One of the crucial point in the present study is the anisotropic dynamical behavior of MOVPE growth. Hersee *et al.*[4] and Bhat *et al.*[5] have reported the different dynamical behavior of MOVPE growth between two types of patterned substrates having grooved feature along the [110] or [1$\bar{1}$0] direction. In fact, as shown in Fig. 2(a), the surface after the growth of GaAs/AlGaAs layered structures is highly anisotropic with appearance of rhombic patterns.

To further investigate this anisotropic behavior, we took cleaved cross-sectional SEM images of the layered structures. The results are shown in Fig. 2(b) and (c), where the direction of the cleavage lays along [1$\bar{1}$0] (X–X′ in Fig. 1(a)) and [110] (Y–Y′ in Fig. 1(a)) directions, respectively. Here, AlGaAs layers are used as markers and correspond to the brighter regions in the figures. One can readily see the highly anisotropic dynamical behavior of the growth. It is also noted that it differs significantly from the case of growth on patterned substrates with grooved features [4, 5], indicating the importance of complex three-dimensional diffusion of Ga and Al atoms between different facets.

Since the growth dynamics produces a bend in Fig. 2(b) and successive size reduction in Fig. 2(c), formation of three-dimensionally confined GaAs dots together with surrounding sidewall and top quantum well is expected as schematically shown in Fig. 3. To

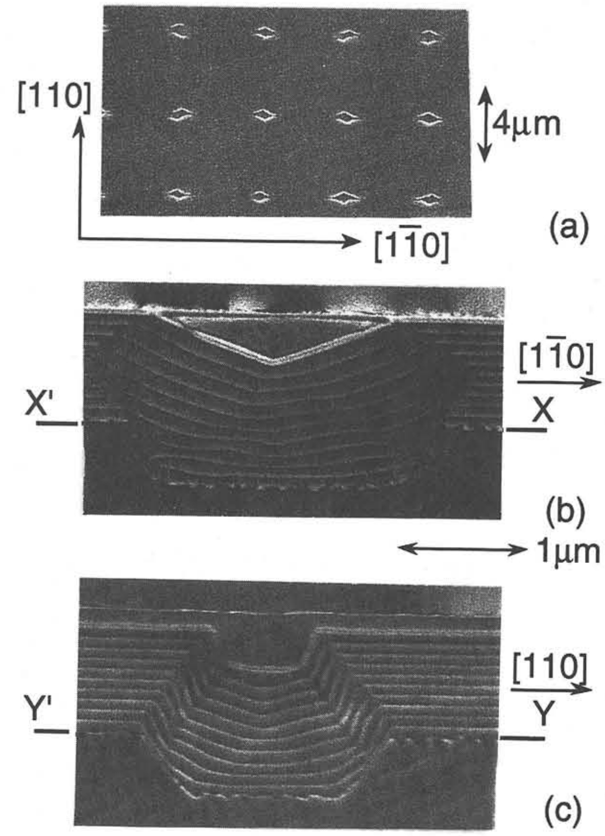

Fig. 2: SEM images taken after the growth of GaAs/AlGaAs layers (a) plane view, (b) cross-sectional view along the [1$\bar{1}$0] direction, and (c) cross-sectional view along the [110] direction.

confirm this expectation, cathodoluminescence (CL) measurements of the sample were carried out at 11K and investigated the luminescence from the well region formed near the top. Figure 4(a) shows the spatially integrated CL spectra. Here, the excitation electron beam was selectively focused only around a rhombic patterned region. Three luminescence peaks originated from the well can be observed. To identify the origin of these peaks, the CL images were taken separately at three different wavelength of 784nm, 800nm, and 805nm corresponding to peaks in Fig.

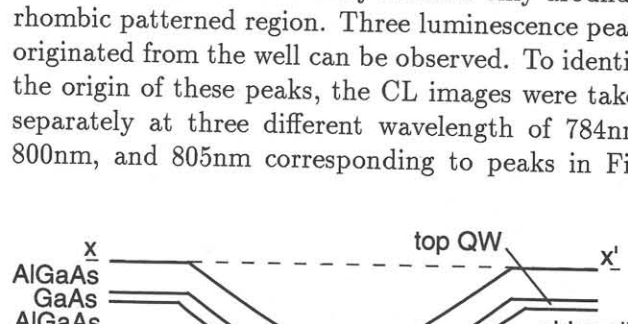

Fig. 3: Schematic representation of the GaAs dot and surrounding quantum wells

4(a), and results are respectively shown in Fig. 4(b), (c), and (d). One can see excellent correspondence between emission peaks and spatial locations. Complementary emission between Figs. 4(b) and (c) shows a clear distinction between the top (001) surface and the rhombic holes. Moreover, brighter regions in Fig. 4(d) correspond to the bottom of the rhombic holes and are clearly distinguished from the sidewalls of the holes. These results clearly indicates the formation of GaAs dots at the bottom of the rhombic holes.

The relative red-shift of the luminescence peak of the sidewall QWs to that of top QW can simply be explained by the difference of the growth rates between two surfaces. In fact, from CL spectrum of Fig. 4(a), the well width of the sidewall QW and top QW is estimated to be 11.2nm and 6.9nm, respectively. This is consistent with the SEM data of Fig. 2(b), where the ratio of the growth rate of the sidewall to that of the top is estimated to be 1.6. On the other hand, the width of the bottom quantum well is estimate to be 14.2nm, discarding the lateral confinement. Such large well width cannot be explained simply by the SEM data and is thought to suggest the accumulation of GaAs at the bottom, as in the case of V-grooved features[5].

Because the lateral size of the dots is still in the range of 0.1μm and the well width is still too large, we have no evidence for lateral quantum confinement at present. It should be noted, however, that significant emission was observed from such small dots. This fact can be ascribed to the presence of smooth carrier transfer from the surrounding sidewall quantum wells, and low defect densities of the present structures.

4 Summary

We have studied the growth dynamics of the MOVPE on GaAs substrate patterned by an array of rectangular holes and applied this technique to the fabrication of quantum dot structures. Strong anisotropic behavior of the growth was found, showing significant difference from the case of wire formation. Spatially resolved cathodoluminescence measurements demonstrated the formation of GaAs dot structures at the bottom of rhombic holes.

Acknowledgments

The authors are grateful to T. Ohba, J. Ishizaki, S. Hara, and K. Nakakoshi for experimental support. This work is partly supported by the Grant in Aid for Scientific Research from the Ministry of Education, Science and Cultures.

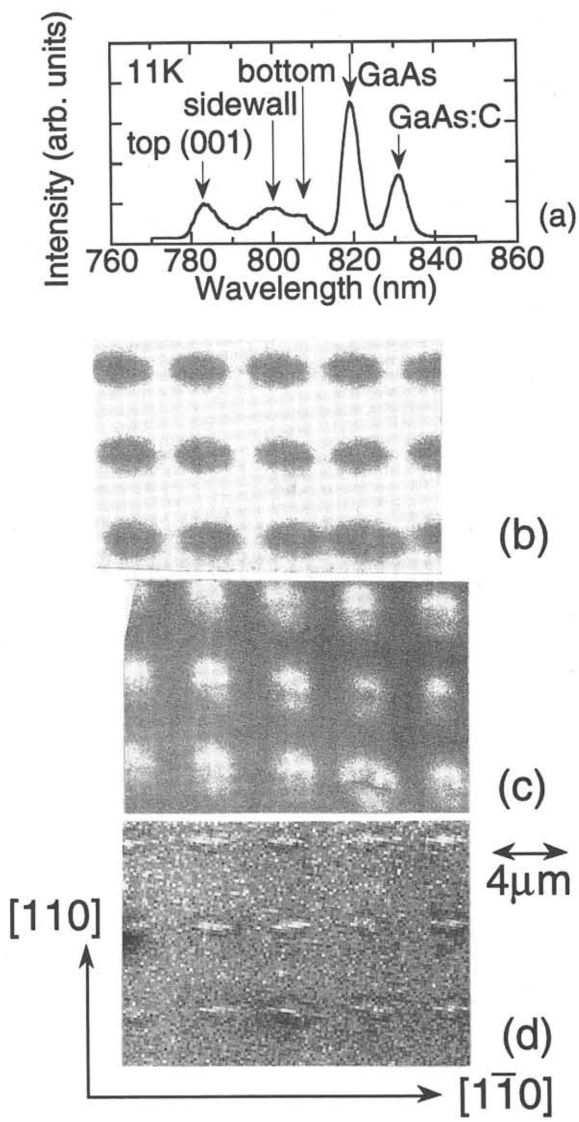

Fig. 4: Spatially integrated CL spectra (a) and two dimensional CL image of the sample taken at fixed wavelength; (a) 784nm, (b) 800nm, and (c) 805nm.

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