MOVPE Condition Dependences of MnAs Nanoclusters Grown on GaInAs (111)A Surfaces

Hiroko Iguchi, Shinjiroh Hara, Junichi Motohisa and Takashi Fukui

Research Center for Integrated Quantum Electronics, Hokkaido University, North 13, West 8, Sapporo 060-8628, JAPAN Phone: +81-11-706-7172, Fax: +81-11-716-6004, e-mail: iguchi@rciqe.hokudai.ac.jp

1. Introduction

Epitaxial hetero-structures consisting of ferromagnetic materials and III-V compound semiconductors have been highly expected for fabricating magnetic tunnel junctions (MTJs) because they can be used to make magneto resistive random access memories (MRAMs). [1] Recently, it has been reported that tunneling magneto resistance (TMR) effect are observed for the MnAs/AlAs/MnAs trilayers even above room temperature. [2] MnAs epitaxial layers can be grown on commonly used semiconductor substrates. For the growth of hexagonal NiAs-type MnAs layers, {111} orientations of zinc-blende-type materials are suitable because of the similarity of atomic arrangements in the crystal structures. We have investigated the formation of MnAs nanoclusters on GaInAs/InP (111)A and B layers by metal-organic vapor phase epitaxy (MOVPE), which is one of the most important and preferred technologies for the fabrications of electronic and photonic semiconductor devices. [3, 4] It was confirmed from the surface observation results that hexagonal MnAs nanoclusters were formed on GaInAs (111)A and B surfaces. Much smaller nanoclusters were formed in higher density on the GaInAs (111)A surfaces than on the GaInAs (111)B surfaces. [4] That experimental results seemed to show the potential possibility to grow MnAs thin films on GaInAs (111)A surfaces. In this paper, therefore, we fundamental MOVPE condition investigate the dependences of ferromagnetic MnAs nanocluster formation on planar GaInAs/InP (111)A layers.

2. Experimental procedures

First of all, GaInAs/InP buffer layers were grown on planar InP (111)A substrates by MOVPE. InP (001) and (111)B substrates were used as a reference at the same growth runs. Growth temperature, Tg, for the growth of InP buffer layer was 600 °C, and V/III ratio was 50. The GaInAs buffer layer was grown under the conditions of $T_g = 650$ °C and V/III = 100. After growing GaInAs/InP buffer layers, MnAs layers were grown. Tg for the MnAs layers was set to 600 °C for all of the samples. The growth times were changed from 10 seconds to 10 minutes to characterize the surface morphologies of the sample with MnAs nanoclusters grown on the GaInAs (111)A surfaces. The V/Mn ratios were changed from 30 to 1125. Here, the supply gas ratio between the partial pressures of a group V source, $p[AsH_3]$, and a manganese precursor, $p[(CH_3C_5H_4)_2Mn]$, is defined as a V/Mn = $p[AsH_3]/p[(CH_3C_5H_4)_2Mn]$, and is referred to

as a "V/Mn ratio". Surface morphologies of the samples were observed by atomic force microscopy (AFM). Cross-sectional lattice images were observed by transmission electron microscopy (TEM).

3. Results and discussion

Initially, we revealed by AFM that GaInAs layers grown on InP buffer layers had no nanoclusters on the surface. (Fig. 1 (a)) Formation of nanoclusters was observed on GaInAs/InP (111)A layers after the MnAs growth, as shown in Fig. 1 (b). The growth time of the MnAs layers for this sample was 1 minute, and V/Mn ratio was 60. The average height and diameter of nanoclusters were 11.4 nm and 250 nm, respectively. The density of the nanoclusters was estimated to be 3.3 x 10^9 cm^{-2} . Fig. 2 shows V/Mn ratio dependences of the MnAs nanocluster formation. The results remarkably show that the size and density of the nanoclusters are increased with increasing V/Mn ratios. Figure 2 (a) also showed that deep holes were observed around the MnAs nanoclusters grown under the low V/Mn ratio condition of 60. These deep holes were never observed for the samples grown on GaInAs (001) and (111)B surfaces.



Fig. 1 AFM images showing the morphologies of (a) the GaInAs (111)A surface and (b) MnAs nanoclusters grown on the GaInAs surface. The observed area was $5 \times 5 \text{ um}^2$.



Fig. 2 V/Mn ratio dependences of the formation of MnAs nanoclusters. Growth time for the MnAs layers was 10 minutes for both of the samples, and the observed area was $5 \times 5 \text{ um}^2$. (a) Deep holes were observed around the nanoclusters (V/Mn = 60), and (b) MnAs nanoclusters were formed in higher density (V/Mn = 750) than on the samples shown in Fig. 2(a).

Next, in order to closely investigate crystallographic relationship between MnAs and GaInAs, cross-sectional TEM images were taken for the samples grown on GaInAs/InP (111)A layers. MnAs nanoclusters embedded into GaInAs (111)A layers were observed, as shown in Fig. 3 (a). In addition, the tilted c-axis of the MnAs nanoclusters with respect to the [111] direction of the GaInAs layers was observed. As shown in Fig. 3 (b). one of the observed nanoclusters was inclined by 70° from the [111] direction, and another was almost parallel to the [111] orientation. Therefore, the deep holes shown in Fig. 2 (a) were not caused by the roughness formed in the MnAs layers, but the one formed on the underlying GaInAs layers. It was reported that similar nanoclusters embedded in the host matrix semiconductor materials were observed in the case of MnSb on GaSb (001) layers grown by molecular beam epitaxy. [5] The phenomenon discussed in Ref. [5] has been understood as an "endotaxy". [6] The deposited materials form oriented single crystals that grow into the substrate materials as a result of "endotaxy". Therefore, the phenomenon observed in our samples is presumably caused by the diffusion of Mn ad-atoms into the underlying GaInAs layers because of the endotaxy. It has been confirmed that higher density of the MnAs nanoclusters are formed on GaInAs (111)B surfaces at larger V/Mn ratios. Furthermore, we have been recently investigating that large V/Mn ratios are needed in the case of selective-area MOVPE on partially SiO₂-masked GaAs (111)B surfaces. Therefore, we possibly conclude that the bonding between As and Mn atoms is promoted under the large V/Mn ratio conditions. Since the (111)A surfaces of zinc-blende-type materials are consisted of group III atoms, much larger V/Mn ratios are needed for the MnAs growth, in particular, on GaInAs (111)A surfaces.

Conclusions

MnAs nanoclusters embedded in GaInAs (111)A layers were observed. The growth of MnAs nanoclusters was enhanced with increasing V/Mn ratios.

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Fig. 3 Cross-sectional TEM images (a) MnAs nanoclusters embedded in GaInAs (111)A layers were observed. (b) C-axis of MnAs nanoclusters tended be titled against the [111] directions of the GaInAs layers.