

**Fabrication and properties of III-V/MnAs hybrid structures
and III-V based ferromagnetic semiconductors**

(III-V/MnAs 複合構造および III-V 族ベース強磁性半導体の作製と物性に関する研究)

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Abstract

This thesis presents the studies on the fabrication and fundamental properties of nanostructure materials for semiconductor spin-based electronics; MnAs thin films, III-V:MnAs granular thin films, and III-V quarternary alloy ferromagnetic semiconductor $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films.

A variety of materials and structures for semiconductor spin-based electronics have been investigated for decades. For example, III-V ferromagnetic semiconductors, such as $(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$ and $(\text{In}_{1-x}\text{Mn}_x)\text{As}$, are potential candidates, but their Curie temperatures are much lower than room temperature. On the other hand, spinodal decomposition which is often observed in wide-gap-semiconductor based diluted magnetic semiconductors and diluted magnetic oxides, such as GaN:Mn and ZnO:Co, has recently generated much attention, because it can lead to high Curie temperature, but such materials have some difficulties for device applications. In this thesis, we have fabricated and investigated the properties of ferromagnetic materials which have high Curie temperature and excellent compatibility with III-V semiconductor devices.

First, the growth and fundamental properties of ferromagnetic MnAs thin films on InP substrates are described. We have successfully grown the MnAs thin films on InP(001) substrates by molecular beam epitaxy (MBE), and characterized the structural and magnetic properties. The epitaxial orientation was as follows: MnAs $(\bar{1}100) // \text{InP}(001)$, MnAs[0001]//InP $[\bar{1}10]$, and MnAs $[11\bar{2}0] // \text{InP}[110]$. The easy magnetization axis was in-plane and along the MnAs $[11\bar{2}0]$ direction. The Curie temperature was 321 K. We also succeeded in the growth of MnAs thin films on $\text{In}_{0.65}\text{Ga}_{0.35}\text{As}_{0.75}\text{P}_{0.25}$ layers in the same manner. Excellent compatibility of MnAs with InP based materials is demonstrated.

Next, the fabrication and fundamental properties of the III-V:MnAs granular thin films, where MnAs nanoclusters are embedded in a host III-V semiconductor matrix, are described. Generally, overgrowth of a semiconductor layer on a ferromagnetic layer is very difficult because of the difference in crystal structure between ferromagnets and semiconductors. However, a semiconductor layer can be easily grown on a III-V:MnAs granular layer, and this is one of the advantages of the granular thin films. We fabricated GaAs:MnAs granular thin

films with MnAs nanoclusters of NiAs-type crystal structure, by annealing $(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$ films at 600°C . GaAs:MnAs granular thin films showed a large magneto-optical effect and a large magnetoresistance change ($>600\%$) at room temperature. Furthermore, by annealing $(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$ around 500°C , we also fabricated GaAs:MnAs granular thin films with MnAs nanoclusters of zinc-blende(ZB)-type crystal structure. The Curie temperature of the GaAs:MnAs granular thin films with ZB-type MnAs nanoclusters was 360 K.

We also succeed in fabricating $(\text{In}_y\text{Ga}_{1-y})\text{As}:\text{MnAs}$ and $(\text{In}_y\text{Al}_{1-y})\text{As}:\text{MnAs}$ granular thin films, which are compatible with InP. These III-V:MnAs granular thin films showed a large magneto-optical effect at room temperature. Adding to the unique fundamental properties, the III-V:MnAs granular thin films were well compatible with both GaAs and InP based III-V semiconductors. Furthermore, we applied the $(\text{In}_y\text{Ga}_{1-y})\text{As}:\text{MnAs}$ and $(\text{In}_y\text{Al}_{1-y})\text{As}:\text{MnAs}$ granular thin films to a magneto-optical device operating at room-temperature, that is, a semiconductor waveguide-type optical isolator based on the nonreciprocal loss effect. We integrated the III-V:MnAs granular layers into semiconductor optical amplifier, and observed the optical absorption based on the nonreciprocal loss shift.

Finally, the fabrication and the fundamental properties of III-V quaternary alloy ferromagnetic semiconductor $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films are presented. In particular, their magnetic anisotropy was investigated. We found that the compressively strained $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films showed uniaxial magnetic anisotropy along the $[\bar{1}10]$ direction by using planar Hall effect measurements. We controlled the magnetic anisotropy of $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ by changing the In content and thus the strain in the ferromagnetic films. A series of the $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films were grown by MBE with various In content (y) of 0.38 – 0.54 and with Mn content (x) of 0.12. As y increased, that is, the compressive strain becomes dominant over the tensile strain, the easy magnetization axis changed to an in-plane direction from the perpendicular direction. The relationship between the strain and magnetic anisotropy in $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ well agreed with that in other III-V ferromagnetic semiconductors ($(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$ and $(\text{In}_{1-x}\text{Mn}_x)\text{As}$). Additionally, we found that the perpendicular component of the magnetic anisotropy is increase by low-temperature annealing, probably because of the reduction of the interstitial Mn atoms. Moreover, we grew the $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films with high x of up to 0.40. When the thickness of $[(\text{In}_y\text{Ga}_{1-y})_{1-x}\text{Mn}_x]\text{As}$ thin films was below 10 nm, the Curie temperature was around 100 K. On the other hand, the thickness was above 10 nm, especially at a thickness of 100 nm and $x = 0.40$, the Curie temperature was above the room temperature, though the origin of high temperature ferromagnetism needs to be further investigated.