Hot-Wall Growth and Magneto-Optical Studies of MnSb Films on GaAs Substrates

H. Ikekame,**** Y. Morishita,* and K. Sato*

*Faculty of Technology, Tokyo University of Agriculture
and Technology, Koganei, Tokyo 184

**Research Fellow of the Japan Society for the Promotion of Science

Ferromagnetic MnSb thin films were grown on GaAs (100) substrates by the hot-wall epitaxy (HWE) technique using polycrystalline powders of MnSb as an evaporation source. The source and substrate temperatures were 650–700°C and 300–500°C, respectively. The (10.1) oriented MnSb films of nearly stoichiometric composition were grown on (100) GaAs. The polar magneto-optical Kerr rotation and ellipticity spectra were measured in the films, and showed almost the same spectral shape as that reported previously in bulk single crystals, except that the magneto-optical effect of the films was larger in the high-energy region than that of the single crystals.

Key words: MnSb, GaAs, epitaxy, hot wall epitaxy, magneto-optical Kerr effect, hybrid magnetic semiconductor

1. Introduction

MnSb intermetallic compound with a nearly stoichiometric composition is a well-known ferromagnetic material, in which magnetic and magneto-optical properties have been intensively investigated from both fundamental and applicational view points. 2), 3)

In our previous study, magneto-optical Kerr spectra of MnSb single crystals for wide photon energies region were investigated,^{4),5)} because only available data on MnSb had been the one measured by Buschow *et al.* on polycrystalline bulk materials, in which spectral region was limited between 1 and 4 eV.²⁾

Recently, novel hybrid structures consisting of semiconductors and magnetic substances are attracting much interest as possible candidate materials for new functionality device application.⁶⁾ For this purpose growth technology of thin magnetic films of a good quality on the semiconductor substrates are strictly needed. Several reports have already been published on the preparation techniques and the properties of magnetic materials on III–V compound semiconductor substrates. For instance, MnAs thin films⁷⁾ and MnGa/ NiGa artificial superlattices⁸⁾ were grown epitaxially on GaAs substrates and so on.

For these hybrid magnetic semiconductor system, MnSb is one of desirable ferromagnetic materials, because it consists of a transition element and a group V element, which make it possible to fabricate the hybrid heterostructures with abrupt interface consisting of Sb-based magnetic materials, without interfacial diffusion which had been seen in Fe grown on GaAs system.⁹⁾

Akinaga *et al.* succeeded in growing epitaxial ferromagnetic MnSb thin films by molecular beam epitaxy technique and reported the orientation dependence of magneto-optical properties.¹⁰⁾ Tatsuoka *et al.* also reported the epitaxial growth of MnSb on GaAs substrate by HWE using Mn and Sb element materials as evaporation sources.¹¹⁾

In the present paper, we report the growth of ferromagnetic MnSb thin films on GaAs substrates by HWE technique from the source of polycrystalline MnSb compounds and the characterization of their magneto-optical properties.

HWE is a very versatile and economical technique suited for basic researches of epitaxial growth condition of new materials. In this technique, 12) the epitaxial growth is carried out under nearly thermal equilibrium condition and a loss of materials can be minimized. The HWE system is mainly composed of three parts; *i.e.*, a hot-wall tube to transport the evaporated materials, a substrate placed at the top of the tube and an evaporation source at the lower end of the tube. Thd role of the hot-wall is to keep the temperature of the vapor uniform. The space enclosed by these parts prevents an out-diffusion of the vapor, which in turn makes the control of vapor pressure easy. Since thermal contacts among these parts are relatively weak, individual change and control is possible in some extent.

2. Experimental Procedure

The GaAs substrates were cut parallel to (100) plane with inclination towards the [011] direction by 2 degrees. The reason why we used off-axis substrate is that easier epitaxial growth has been reported than using the just (100) GaAs substrates. The substrate was set on a molybdenum holder with indium contacts in the HWE system. The chamber was then evacuated to the pressure of 10^{-7} Torr, in which thermal cleaning of the

substrate was carried out at 580°C for 5 minutes.

Thin films of MnSb were grown on the (100) surface of GaAs substrates without any buffer layer. As the evaporation source, polycrystalline MnSb compound synthesized from elements of Mn and Sb by the normal freezing technique was used, the details having been described in our previous paper.⁴⁾

In the present investigation, growth conditions are as follows; the source and substrate temperatures were varied in the range of 650–700°C and 300–500°C, respectively. The growth rates were maintained about 0.1–0.2Å/s, and the typical thickness of the thin films obtained was about 2000Å.

Characterization of films were carried out as follows; structural properties were studied by XRD technique using a Rigaku RAD-IIc diffractometer (Cu- K_a). The composition of MnSb thin films were analyzed by means of EPMA using a JEOL JXA-8900R microanalyzer. The surface morphologies were observed using a JEOL JSM-5200 scanning electron microscope (SEM). The polar magneto-optical Kerr rotation and Kerr ellipticity spectra were measured at room temperature for photon energies region from 1.2 eV to 5.8 eV using a specially designed Kerr spectrometer developed in our laboratory.

3. Results and Discussion

A typical XRD pattern observed in the MnSb thin film is shown in Fig. 1 ($T_{\rm source} = 700^{\circ}\text{C}$ and $T_{\rm substrate} = 450^{\circ}\text{C}$). Only diffraction lines indexed as 10.1 and 20.2 are observed in addition to the lines related to the GaAs substrate. No other diffraction lines were observed. This suggests an epitaxial growth of MnSb with the (10.1) plane of hexagonal NiAs structure parallel to the (100) plane of GaAs substrate. The same epitaxial relationship was also reported in previous studies. 10 , 11

Similar XRD patterns were obtained in all the samples prepared in this study regardless of the difference in the growth conditions. This suggests that HWE technique is suited for the growth from the compound source, since it is known that other techniques starting from element sources often lead to a formation of extraneous phases when the growth conditions are changed in wide range.

Table 1 shows the full width at half maximum (FWHM) of the 10.1 XRD line in the films prepared under different conditions. The values of FWHM are no more than 0.24 deg. The average of the FWHM is 0.2 deg. corresponding to the X-ray coherent length of about 300Å, and should be compared with the total film thickness, *i.e.*, 2000Å. The optimum condition was $T_{\rm source}$ of 700°C and $T_{\rm substrate}$ of 450°C. Systematic change of FWHM with the growth condition could not be observed.

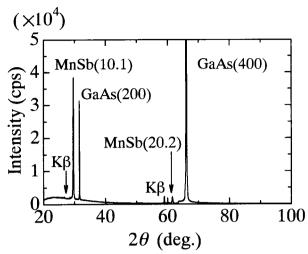


Fig. 1 Typical XRD pattern of MnSb films grown on (100) GaAs substrates ($T_{\text{source}} = 700^{\circ}\text{C}$; $T_{\text{substrate}} = 450^{\circ}\text{C}$).

Table 1 FWHM values of 10.1 diffraction lines in MnSb films

T substrate	T source (°C)		
(°C)	650	675	700
300		0.212	
350		0.235	0.212
400		0.212	0.188
450	0.188	0.188	0.165
500			0.188

The EPMA result provided a stoichiometric composition within the experimental error (~ 3 at%) in all the films prepared in this study. This means that composition of the film does not depend on the growth condition in our HWE technique. We believe that in our system MnSb is transported from the source to the substrate as the compound form without decomposition.

Figures 2 and 3 show SEM images of the MnSb films prepared with $T_{\rm substrate}$ of 400°C and 350°C, respectively, the source temperatures of both samples being 700°C. The film prepared at higher temperature shows a feature typical to the three-dimensional island growth, while that prepared at the lower temperature shows a streaky morphology along the $[01\overline{1}]$ direction. This corresponds to the fact that the step of the substrate is extending along the $[01\overline{1}]$ direction reflecting the inclination of the substrate to the [011] direction.

It is elucidated that films of a good quality can be obtained at the Tsubstrate as low as 350°C, although the FWHM obtained from XRD is not optimized at this condition. Discrepancy between the XRD results and



Fig. 2 SEM image of the surface of an MnSb film grown at a T_{source} of 700°C and $T_{\text{substrate}}$ of 350°C.

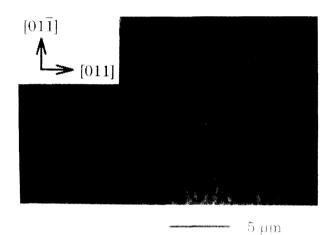


Fig. 3 SEM image of the surface of an MnSb film grown at a T_{source} of 700°C and $T_{\text{substrate}}$ of 300°C.

morphological observation is known to occur also in the epitaxial growth of III-V materials.

The Kerr hysteresis curve showed a magnetic saturation for magnetic field greater than 1 T. Typical polar magneto-optical Kerr rotation and ellipticity spectra of MnSb films are shown in Fig. 4 (The sample was prepared at $T_{\text{source}} = 700^{\circ}\text{C}$ and $T_{\text{substrate}} = 350^{\circ}\text{C}$). The Kerr rotation crosses zero at 4.8 eV where the Kerr ellipticity takes a maximum value. The fact that Kramers-Kronig relationship holds between the rotation and the ellipticity guarantees the high reliability of the measurement. For comparison the Kerr spectra in the bulk single crystal of MnSb reported previously4) is presented in Fig. 5. By comparison it is found that spectral shape of the film was almost the same as that of the bulk single crystal except for the difference in the higher energy region where the ellipticity takes a larger value and the rotation shows a steeper slope than those in the bulk sample. Improvement of Kerr effect in higher energy

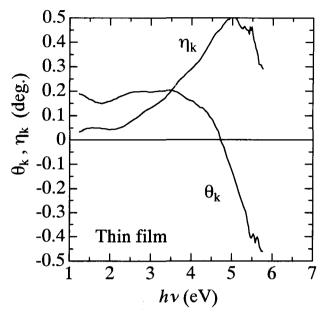


Fig. 4 Typical spectra of polar magneto-optical Kerr rotation and ellipticity in the MnSb films ($T_{\text{source}} = 700^{\circ}\text{C}$; $T_{\text{substrate}} = 300^{\circ}\text{C}$).

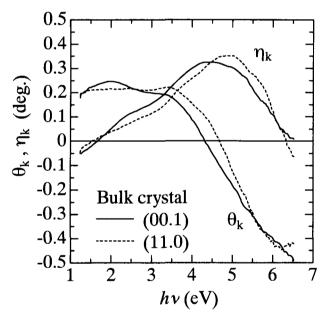


Fig. 5 Spectra of polar magneto-optical Kerr rotation and ellipticity in MnSb single crystals for two crystal planes.

region may be ascribed to the reduced scattering in the surface of the epitaxial film compared with the polished surface of the bulk crystal. Analysis of the magneto-optical spectra in terms of conductivity tensor is now under studies and will be reported in later publications

4. Conclusion

MnSb thin films were grown on (100) GaAs substrates from the evaporation source of polycrystalline MnSb compounds by HWE technique. The XRD patterns in all the samples prepared in this study show the (10.1) oriented growth of MnSb films on the (100) GaAs substrate regardless of the difference in the growth conditions. The composition of the MnSb films analyzed by EPMA was stoichiometric for all samples investigated. The SEM image of the MnSb film prepared at a suitable growth condition showed streaky morphology corresponding to the step of the GaAs substrate. The polar magneto-optical Kerr spectra in the films showed almost the same spectral shape as that of the bulk single crystal except for the improvements in the films for higher energy region.

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