

MnBi Thin Films Prepared by Molecular-Beam Epitaxy Technique

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The Epitaxial growth of MnBi films has been studied by means of molecular beam epitaxy (MBE) technique. Single crystals of MgO (001) and Al₂O₃ (00.1) were employed as substrates. Without any buffer layers, MnBi thin films grown on these substrates were polycrystalline. We then prepared MnBi films using either a Au or a Cu layer as a buffer on the substrates. Using Au buffer layers (10.2)-oriented MnBi films were obtained, yet the magnetic property of these films are poor. On the other hand, using Cu (111) buffer layers MnBi films with a sufficient flatness and better magnetic property were obtained, although some trace of (10.2)-orientated grains was observed.

Keywords: thin film, metal buffer layers, magnetic material, M-H loop, manganese bismuth, molecular beam epitaxy

1. Introduction

The physical properties of MnBi films have been intensively studied in the 1960s, as promising materials for magneto-optical (MO) applications. As is well known, these films have a large intrinsic Kerr rotation

as large as 0.7° at room temperature and a perpendicular magnetic anisotropy, both of which are desirable for MO recording.¹⁻³⁾ However, their large grain size and the existence of several structural phases are disadvantageous.²⁾ The former has caused medium noises for replay signals. The latter is concerned with the crystallographic phase transition from the ferromagnetic low-temperature phase (LTP) to the paramagnetic quenched high-temperature phase (QHT) at approximately 628K, which is close to the Curie temperature of the LTP. This implies the partial erasure of information stored by thermo-magnetic writing.³⁾

In recent years many efforts have been made to improve the magnetic and magneto-optical parameters of the MnBi system by addition of different atoms. In particular, the extensive studies of (Al, Si)-doped MnBi films reinforce their possible applicability for information storage, although the results are still strongly dependent on the preparation conditions. On the other hand MnBi films are prepared on quartz and (001) orientated GaAs substrates by molecular beam epitaxy in ultrahigh vacuum environment.⁴⁾ The results indicated successful epitaxial growth of MnBi films with unprecedented magnetic properties.⁵⁾

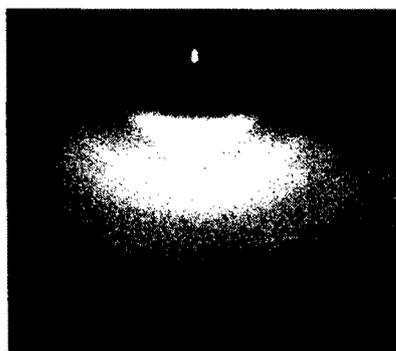
In order to control crystallographic and magnetic properties of MnBi, superlattice structures with other materials such as MnBi/Cu should be investigated. For this purpose growth of MnBi films should be controlled atomically. We therefore studied an epitaxial growth of MnBi by using a molecular beam epitaxy (MBE) technique.

2. Experiments

MnBi thin films were prepared on MgO (001) and Al₂O₃ (00.1) substrates by the MBE method. The Au buffer layer of 1000Å in thickness was evaporated on the substrate in a separate chamber and transferred to



(a) MnBi on MgO(001)



(b) MnBi on Al₂O₃(00.1)

Fig.1 RHEED pattern of MnBi on substrates

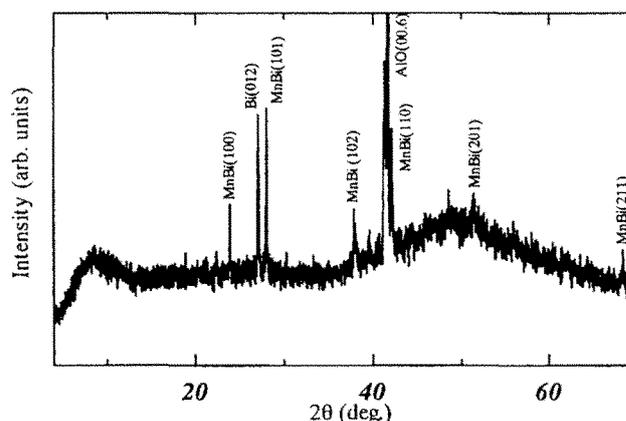
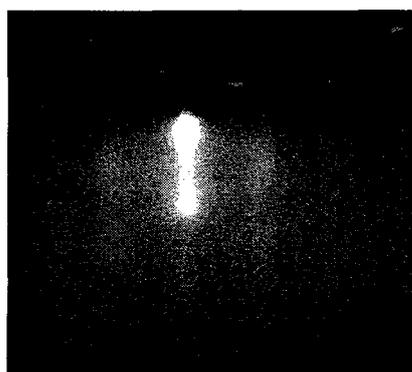
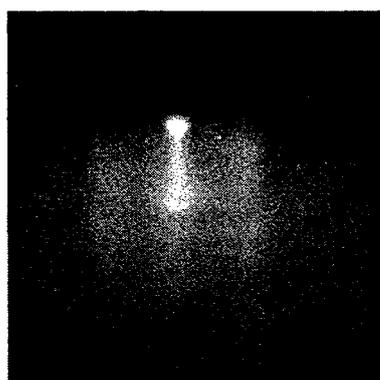


Fig.2 XRD pattern of MnBi on Al₂O₃



(a) Au buffer layers



(b) MnBi layers

Fig.3 RHEED pattern of MnBi on Au/MgO

the MBE chamber, in which it was annealed at 600°C in 3×10^{-8} Torr prior to the growth of MnBi films. On the other hand the Cu buffer layer of 250Å in thickness was deposited in the MBE chamber around 400°C, following the deposition of Cu buffer layer of 1000Å in thickness at 150°C. The Mn and the Bi were co-evaporated on these substrates and these buffer at 300°C. A Cu cap layer of approximately 200Å in thickness was deposited on the MnBi for protection. The structure of the MnBi films was investigated by means of the *in-situ* reflection high-energy electron diffraction (RHEED) and *ex-situ* X-ray diffraction (XRD) measurements. Chemical composition of thin films was measured by electron probe microanalysis (EPMA). Magnetization was determined with a vibrating sample magnetometer (VSM).

3. Results and Discussion

3.1 MnBi on MgO(001) and Al₂O₃

Figure 1(a) shows a RHEED pattern of MnBi thin films prepared on MgO (001) substrates with electron beam incident at [100] MgO azimuth. The ring pattern indicating that the film was polycrystalline was observed. The RHEED pattern illustrated in Fig. 1(b) also shows a ring pattern in the film prepared on Al₂O₃ (00.1) substrates. An XRD pattern of the latter film is given in Fig. 2, in which no diffraction lines other than those for the MnBi phase were found. An Epitaxial

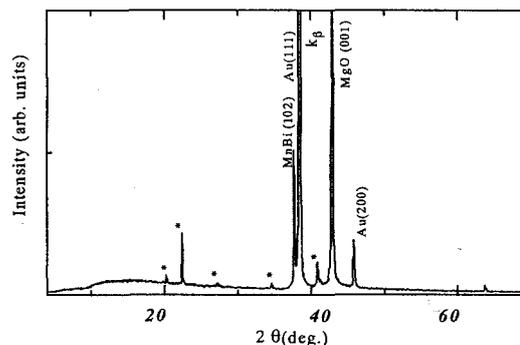


Fig.4 XRD pattern of MnBi on Au/MgO

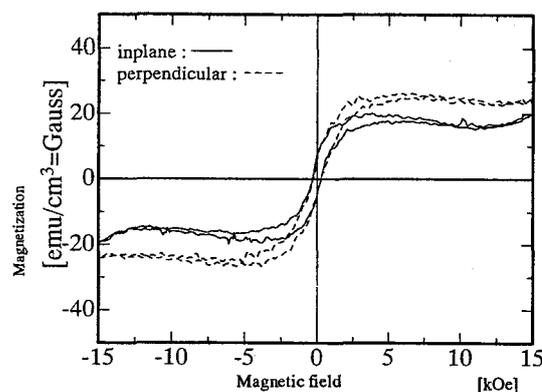


Fig.5 M-H loop of MnBi on Au/MgO

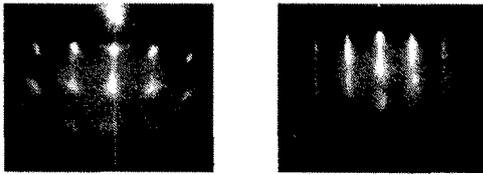
growth of MnBi on these substrates seems to be difficult presumably because of the large lattice mismatch amounting to 10%.

3.2 Growth of MnBi on Au buffer layer

Figure 3 shows RHEED patterns of (a) the Au buffer layer prepared on MgO (001) substrates and (b) the MnBi film on the Au-covered MgO (001) observed with an incident beam along the the [100] MgO axis. The observed pattern of the Au buffer was spotty streak, from which the lattice spacing in the Au buffer was calculated to be 2.808Å. The value is consistent with the theoretical lattice spacing of Au (111). The pattern of the MnBi deposited on the Au buffer was also spotty streak. The lattice spacing calculated was as large as 4.14 Å consistent with the spacing of MnBi (10.2) obtained a literature⁶.

Figure 4 shows an XRD pattern of the MnBi film on the Au/MgO (001), in which we observe diffraction lines due to MnBi (10.2), Au (111) and Au (200). This indicates a growth of a (10.2)-orientated MnBi layer on Au(111). However, the film was not of a single phase, since diffraction peaks due to Mn_xBi_{1-x} phase were clearly observed as marked by asterisks.

Figure 5 shows in-plane and perpendicular M-H loops for MnBi films prepared on Au/MgO (100). The saturation magnetization was nearly one order of magnitude smaller than the typical value. Use of the Au-buffer results in only a partial success.



(a) Cu on MgO(001) (b) Cu on MgO(111)

Fig.6 RHEED pattern of Cu buffer layer

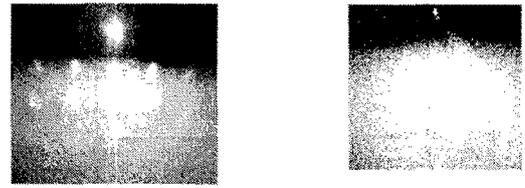


Fig. 7 (a) MnBi on Cu/MgO(001)

Fig. 7 (b) MnBi on Cu/MgO(111)

Fig. 7 RHEED pattern of MnBi on Cu/MgO(111)

3.3 Growth of MnBi on Cu buffer layer

Figures 6 show RHEED patterns of the Cu buffer layer on (a) MgO (001) and (b) MgO (111) observed with the electron beam incident along [100] MgO and [110] MgO azimuth, respectively. The Cu buffer layer on MgO (001) shows a spotty RHEED pattern, indicating that the Cu undergoes a three dimensional growth. On the contrary a nearly streak one was shown on the MgO (111) substrate. This indicates a flatness of the Cu surface obtained. XRD measurements revealed that the orientation of Cu was (001) on MgO (001), while it was (111) on MgO (111).

MnBi thin films were prepared those Cu buffer layers. The intensity of the RHEED pattern from the MnBi films on the Cu/MgO substrates gradually decreased with the deposition time. Figure 7 show RHEED patterns of MnBi films prepared on (a) Cu/MgO (001) and (b) Cu/MgO (111), respectively. The former pattern is clear since it was observed at 30 s after the start of deposition, while the latter is obscured since it was observed at 3 min after the start. The pattern in Fig. 7(a) is considerably different from that expected for MnBi. The lattice spacing is as small as 3.33 Å, which is associated with MnCuBi phases. On the contrary the film on Cu/MgO (111) shows a streak pattern showing that the MnBi has flat terraces.

XRD patterns of MnBi films on Cu/MgO (001) and Cu/MgO (111) are shown in Figs. 8(a) and 8(b), respectively. Fig. 8(a) clearly shows diffraction lines due to Cu (001), MnCuBi (222) and MnCuBi (004), which indicate that the MnCuBi on MgO (001) is polycrystalline. Katui reported the similar XRD pattern previously.⁷⁾ On the other hand, diffraction lines due to Cu (111), MnBi (11.0) and MnBi (10.2) were observed in XRD pattern of the film prepared on Cu/MgO (111) as shown in Fig. 8(b). No diffraction lines due to MnCuBi were observed in this case. MnBi films consisting of grains with flat terrace oriented in (10.2) and (11.0) were grown on Cu (111). The lattice parameter was slightly different from the theoretical value. The discrepancy may be attributed to an incorporation of Cu atoms in the MnBi layers.

Figures 9(a) and 9(b) are M-H loops of MnBi film on Cu/MgO (001) and Cu/MgO (111), respectively. Solid curves were measured parallel to the plane, while dotted ones perpendicular to the plane. Figure 9(a) shows that the magnetic moments in the MnBi film on Cu/MgO (001) has both in-plane and perpendicular components. On the contrary M-H loops of MnBi on

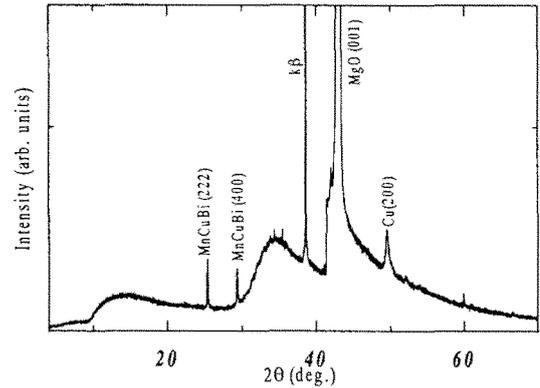


Fig.8 (a) XRD pattern of MnBi on Cu/MgO(001)

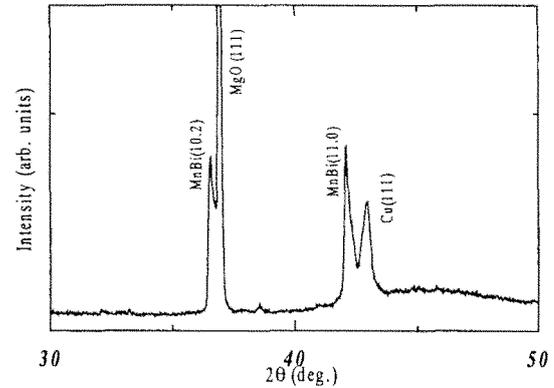


Fig.8 (b) XRD pattern of MnBi on Cu/MgO(111)

Cu/MgO (111) shown in Fig. 9(b) clearly indicate that the film has a perpendicular anisotropy. The saturation magnetization was approximately one half of the typical value.

The room temperature magneto-optical Kerr loops measured at the wavelength of 632.8 nm in MnBi films on the Cu/MgO (001) and Cu/MgO (111) substrates are presented in Figs. 10(a) and 10(b), respectively. The value of the Kerr rotation is larger in the latter film than the former, indicating that the magnetic property in the MnBi film on Cu/MgO (111) is much better than that on Cu/MgO (001). Nevertheless the Kerr rotation was no more than 0.15°, which is less than one thirds of the literature value. The Smallness of the magneto-optical

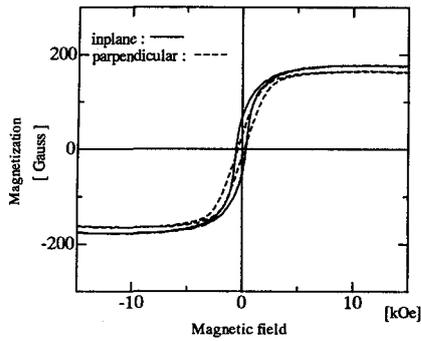


Fig.9 (a) M-H loop of MnBi on Cu/MgO(001)

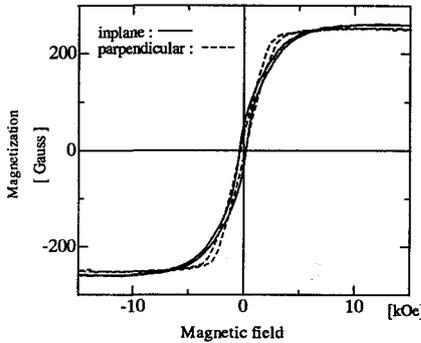


Fig.9 (b) M-H loop of MnBi on Cu/MgO(111)

effect may partly be attributed to the Cu cap-layer deposited on the MnBi film as a protection layer.

Further studies are necessary to improve the magnetic and magneto-optical properties of the material.

4. Conclusion

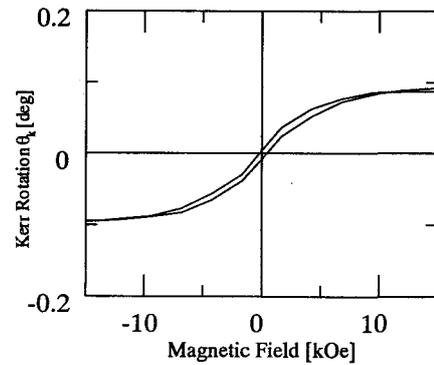
MnBi films have been prepared by means of the MBE technique. Polycrystalline MnBi thin films were grown on these substrates without buffer layers. Using Au buffer layers (10.2)-oriented films of MnBi were obtained. Only a poor magnetic property was observed in these films. On the other hand, using Cu (111) buffer layers so flat as showing a streak RHEED, (11.0)-oriented MnBi films with improved magnetic and magneto-optical properties were obtained, although some trace of (10.2)-orientation were still observed.

Acknowledgements

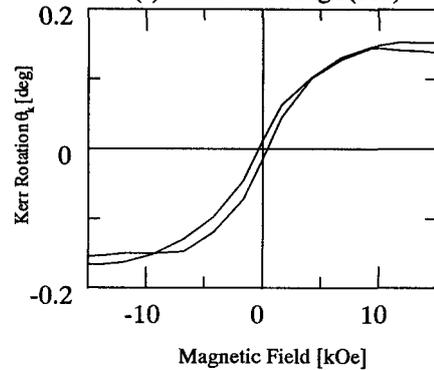
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(a) MnBi on Cu/MgO(001)



(a) MnBi on Cu/MgO(001)

Fig.10 Kerr loops of MnBi on Cu/MgO(001) and Cu/MgO(111)

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