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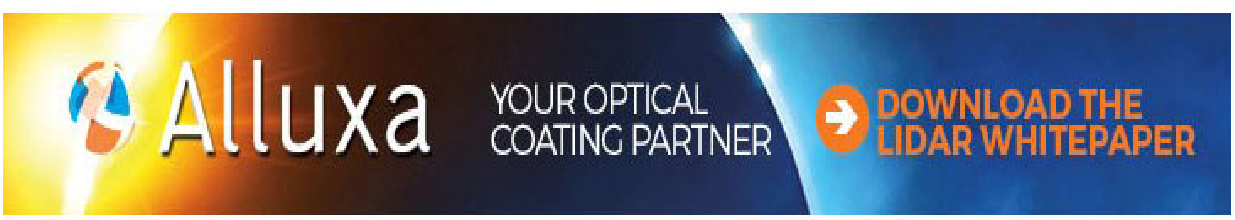

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Magnetic and structural properties of MnBi multilayered thin films

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Magnetic and structural properties of MnBi films with thicknesses up to 50 nm were investigated. Thin films of the MnBi LTP (Low Temperature Phase) were fabricated onto silica-glass substrates by sputter-deposition of Bi/Mn multilayer, followed by a subsequent annealing at about 550 °C for 30 min. Coercivity of such thin films is higher than 15 kOe, even though the film thickness is about 10 nm. These thin films show the preferential growth of *c*-axis of the LTP along the film normal. Moreover, high resolution transmission electron microscopy indicates that the LTP regions of 30–50 nm in size are physically isolated by Bi. The magnetization reversal mechanism of such a LTP region is mainly governed by a coherent rotation mode based on the δM curve measurement.
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I. INTRODUCTION

Thin film permanent magnets have received much attention because of potential applications for MEMS.[1,2] Among many thin film magnets, intermetallic compound MnBi with the low temperature phase (LTP) is of great interest because of a large magnetic anisotropy constant (K) of the order of 10^7 erg/cc,[3] which increases with temperature (T) over a temperature range from about 200 K to about 600 K and also because it does not contain high-cost elements such as rare-earths.

In view of the above mentioned, much work of the LTP MnBi thin films has been carried out.[4–6] However, there has been little work found in literature regarding the detailed correlation between magnetism and structure in very thin films with thicknesses of about a few tens nm. The present study has been systematically carried out to understand such a correlation in MnBi thin films.

II. EXPERIMENTAL

Multilayers of (Bi/Mn) $\times$ N (N = 1–10) were fabricated by sputter-deposition onto silica-glass substrates at an ambient temperature during deposition. The deposition rates for Bi and Mn were 0.071 nm/s and 0.017 nm/s, respectively. After preliminary experiments for optimization of fabrication condition, the thicknesses of each Bi and Mn layer were chosen to be 3.2 nm and 2.0 nm, respectively. The multilayers thus deposited were annealed in vacuum at about 550 °C for 30 min.

The crystal structure and microstructure of films were characterized by X-ray diffraction (XRD) with Cu $K\alpha$ radiation and by transmission electron microscopy (TEM). Measurements of the magnetic properties were carried out by using an alternating gradient magnetometer (AGM) in fields up to 18 kOe and by using a vibrating sample magnetometer (VSM) in fields up to 33 kOe.

III. RESULTS AND DISCUSSION

Figure 1 shows a series of XRD spectra for all the films. The diffraction peaks for N = 5, 7, and 10 are clearly assigned to be those for the LTP MnBi, except for N = 10 where a diffraction peak assigned to Bi was also observed. It is noted that the quenched high temperature phase (QHTP) is absent, though one would expect it from the bulk phase diagram,[7] and the previous papers of thick films were reported to consist of both the LTP and QHTP.[8,9] The relatively strong peaks of (002) and (004) indicate a preferential epitaxial growth of the *c*-axis normal to the film plane. However, the presence of the weak intensity of (102) and

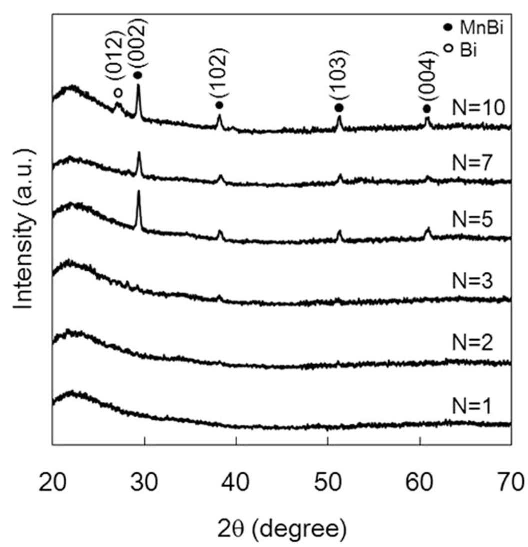

FIG. 1. X-ray diffraction patterns of MnBi films with repeat numbers N of 1, 2, 3, 5, 7, and 10.

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(103) peaks indicates that the LTP with other orientation is also grown in those films. The lattice constants *c* were estimated to be 6.08 Å (N = 5), 6.07 Å (N = 7), and 6.09 Å (N = 10). These values are smaller by −0.75% (N = 5), −0.91% (N = 7), and −0.59% (N = 10) than that of the bulk of 6.126 Å.[10] For those with N < 5, there is little evidence of diffraction lines. However, it may be said, based on the magnetic properties as shown below that ferromagnetic MnBi phase exists for those with N < 5. It is of interest to note that the preferential growth of the *c*-axis along the film normal in such very thin films of MnBi is grown on amorphous silica-glass substrates when a Bi layer is first deposited onto a substrate.

Magnetic hysteresis loops are shown in Fig. 2. In the sample with N = 1, soft magnetic behavior was observed. A similar behavior was also found in the magnetization curves for N = 2. All the magnetization curves for the samples with N ≥ 2 indicate that the easy axis for magnetization is along the film normal. This result is consistent with XRD data, because the magnetic easy axis of the LTP is parallel to the *c*-axis.[11] Coercivity H_c for N ≥ 2 is higher than 15 kOe at room temperature. The present result may be the first demonstration of high H_c in such a very thin MnBi film. One may also conjecture, based on the hard axis slope for magnetization for the samples with N ≥ 2 that the magnetic anisotropy field H_k is about 30–40 kOe, corresponding to a magnetic anisotropy constant K of the order of 10^7 erg/cc.

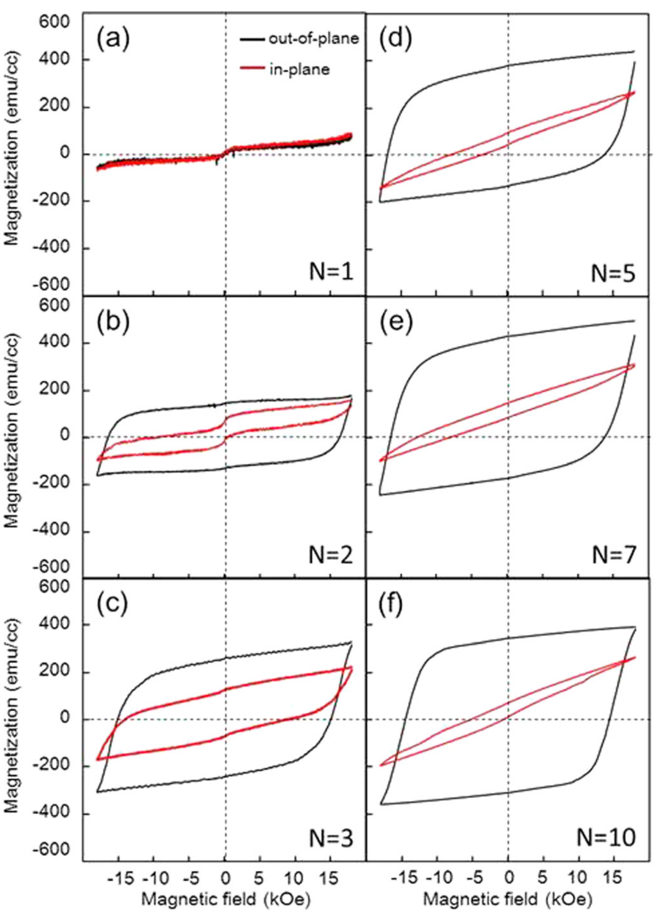

FIG. 2. Out-of-plane (black line) and in-plane (red line) magnetic hysteresis loops for films with N = 1 (a), 2 (b), 3 (c), 5 (d), 7 (e), and 10 (f).

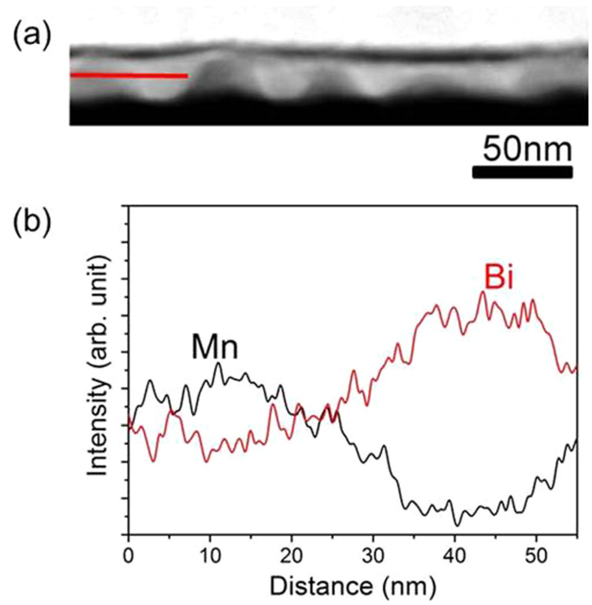

FIG. 3. Cross sectional TEM photograph of the film with N = 10. (a) HAADF-STEM image, (b) EDS intensity profile of Mn and Bi along the line shown in (a).

The microstructure of a cross section of the film with N = 10 was examined by TEM, as shown in Fig. 3. Regions with bright and dark contrast were observed in a High-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image (Fig. 3(a)). From electron diffraction pattern analyses, the bright and dark regions were assigned to the Bi and LTP, respectively. An energy dispersive X-ray spectroscopy (EDS) line profile also showed that the dark region corresponds to that including both Mn and Bi elements to form the LTP region and the bright does to Bi, respectively (Fig. 3(b)). From these analyses, the LTP regions are found to be 30–50 nm in size, which are physically isolated by Bi.

Magnetic interactions among ferromagnetic grains play an important role in a magnetization reversal mechanism. Hence, to understand the magnetic interaction among the LTP regions is important. In the present experiment, measurements of the so-called δM curves were carried out for the film with N = 10. The δM is defined as $\{2M_r(H) - 1 + M_d(H)\}$, where $M_r(H)$ and $M_d(H)$ are the isothermal remanence (IRM) and dc-demagnetization remanence (DCD) curves (normalized to saturation magnetization), respectively.[12] As shown in Fig. 4, the δM curve in the out-of-plane measurement exhibits a negative peak, indicating magnetostatic interaction between the LTP regions is dominant over exchange interaction. This result is consistent with the observation by TEM and EDS, as the LTP regions are physically separated by Bi regions, roughly 20–50 nm apart.

In a spherical particle model, the critical diameter for a single-domain state may be calculated with the following formula:

$$\mathrm{Dc} = \frac{9\varepsilon_w}{2\pi M_s{}^2},$$

where ε_w is the domain wall energy ($\varepsilon_w = 4\sqrt{AK}$, where A is the exchange stiffness constant) and M_s is the saturation magnetization.[13]

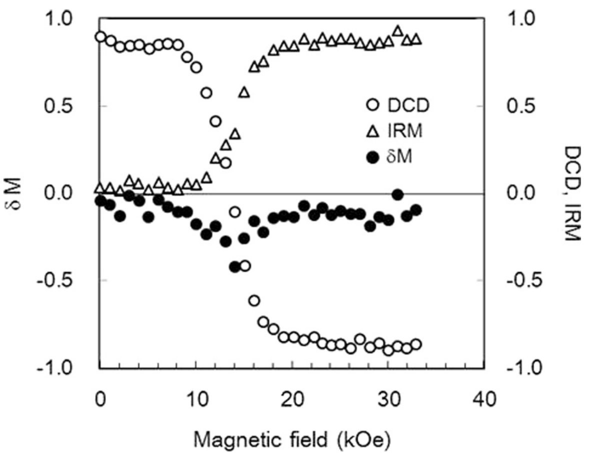

FIG. 4. δM curve (scale on the left) together with IRM (right) and DCD (right) for the out-of-plane measurement for the film with N = 10.

In MnBi, assuming that $M_s = 670$ emu/cc, $K = 1.6 \times 10^7$ erg/cc, and $A = 1 \times 10^{-6}$ erg/cm,[3] the critical diameter is estimated to be about 500 nm. Therefore, the size of an observed LTP region is much smaller than the theoretical critical diameter. Therefore, the magnetization reversal mechanism of the LTP region is believed to be mainly governed by a coherent rotation, leading to a high coercivity. However, since the measured coercivity (about 15–20 kOe) is still smaller than an estimated magnetic anisotropy field of 30 kOe, a contribution of other mechanisms such as domain wall nucleation may not be insignificant.

In summary, MnBi thin films with thicknesses up to 50 nm were fabricated by sputter-deposition of Bi/Mn multilayer and subsequent annealing. Preferential growth of the *c*-axis along the film normal in the LTP MnBi samples is successfully realized. Those films exhibit high H_c higher than 15 kOe. The isolated LTP regions of about 30–50 nm in size are found by TEM/EDS. This size is much smaller than the theoretical critical diameter for a single domain. Therefore, high H_c is believed to result from a coherent rotation mode for the LTP regions. The magnetic anisotropy constant K of such MnBi films is estimated to be 1×10^7 erg/cc.

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