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Magnetic properties and structure of low temperature phase MnBi with island structure

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The magnetic properties of the low temperature phase (LTP) MnBi thin films of islands structure are discussed. The LTP MnBi islands are formed onto silica substrates after the multilayers {Bi(3.2nm)/Mn(2nm)}x N are deposited and then annealed at 450C for 0.5hr, where N is the number of the repetition of a pair of Mn and Bi layer. Those islands are found to be of the LTP MnBi, with the c-axis orientation along the normal to the sample plane for N=10 ∼ 40. Their size vary from place to place, but are averagely of about a few hundred nm in height and a few µm in width for N from 10 to 40. For N=200, the elongated islands are formed densely, with the length of about a few tens of µm. The coverage of those islands increases with N. The temperature dependence of saturation magnetization M_s is qualitatively similar to that for bulk, though the absolute values for M_s are smaller by 20%. The magnetic anisotropy constants of K_{u1} and K_{u2} are evaluated for the samples with N=10 ∼ 40, where K_{u1} and K_{u2} are the magnetic anisotropy constants corresponding to the second and fourth power term in the uniaxial magnetic anisotropy energy expression. It is found that the K_{u1} increases with T monotonously, reaching to about $1 x 10^7$ erg/cc at 400K. On the other hand, the K_{u2} remains nearly zero for temperatures below 300K, and then becomes negative, reaching to about 7 x 10^6 erg/cc at 400K. This is the first to report of the temperature dependence of K_{u1} and K_{u2} in the LTP MnBi of an island structure. It is also noted that the decrease of K_u for a temperature range beyond around 450 K is possibly due to the decrease of the K_{u2} component, as demonstrated in the present study. [http://dx.doi.org/10.1063/1.4977230]

I. INTRODUCTION

Voluminous works have been carried out on the magnetic properties of the low-temperature phase (LTP) MnBi in the forms of bulk, thin films and powders in conjunction with their structures and morphologies.[1–4] In particular, the unique temperature dependence of magnetocrystalline anisotropy constant K_u, that increases with temperature T in the order of 10^7erg/cc for temperatures above about 200K, is the subject for many years.[5–8] Yet, despite many works, there is still a lack of understanding of the magnetic anisotropy mechanism in conjunction with the structure and morphology of the samples investigated. A recent theoretical work[9] based on the first principle calculation emphasized

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the important role of the lattice constant a in the magnetic anisotropy mechanism in LTP MnBi. However, the theory has failed to account for the experimental result of the relation between K_u and T if one used the experimentally obtained lattice constant values of a and c to explain the change of K_u with a and c. Similarly, another recent work[10] on LTP MnBi of island structure showed the discrepancy between the theory and experiment. It was also pointed out that the measured dependence of K_u on M_s over a wide temperature range from 5 to 400K showed that the K_u is inversely proportional to the n^{th} power of M_s, where n is 5 ~ 11.[10,11]

In this present work, in order to elucidate further the temperature dependence of magnetic anisotropy of the LTP MnBi, a systematic experimental study has been performed for various total thicknesses of the samples. In particular, the temperature dependence of both the first and second magnetic anisotropy constants, K_{u1} and K_{u2} are discussed.

II. EXPERIMENTAL

Multilayers of {Bi(3.2nm)/Mn(2.0nm)}N were fabricated onto silica glass by using DC-magnetron sputtering at ambient temperature, where N was varied from 10 to 200. These multilayers thus fabricated were annealed in vacuum at 450°C for 0.5h. The deposition rates for Bi and Mn were about 0.07nm/sec and 0.02nm/sec, respectively. The base pressure of the vacuum chamber prior to deposition was better than 10^{-8} Torr. The detail description of the film fabrication is found elsewhere.[8]

Structural analyses were conducted by X-ray diffraction (XRD) with Cu$K\alpha$ radiation, high resolution transmission electron microscopy (TEM) and by scanning electron microscopy (SEM). Measurements of magnetic properties were carried out by a vibrating sample magnetometer (VSM) in fields up to 90kOe at various temperatures from 5K to 400K. The saturation magnetization M_s was determined by the values of magnetization at 90kOe. The magnetic anisotropy constants K_u, K_{u1} and K_{u2} were evaluated by using a torque method over a temperature range from 5 to 400K in fields up to 90 kOe.

III. RESULT AND DISCUSSIONS

Figure 1 shows the XRD patterns for the {Bi(3.2nm)/Mn(2.0nm)}N films annealed at 450°C for 0.5hr. ((a) N=10, (b) 20, (c) 30, (d) 40, and (e) 200, respectively.) The XRD patterns of the films for N=10~40 indicate that they are of the LTP MnBi with the c-axis orientation of the LTP MnBi along the film normal. It is noted that the samples with N=20, 30 and 40 exhibit the peaks of (101) and (102) of the LTP MnBi, which are of relatively weak intensities. In Fig. 1 (e), the samples with N=200 is

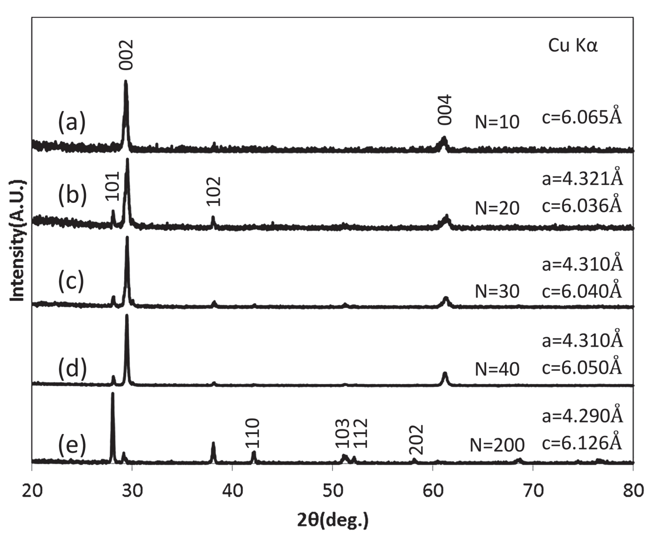

FIG. 1. X-ray diffraction patterns of MnBi samples with the repetition number N: (a) 10, (b) 20, (c) 30, (d) 40 and (e) 200.

not oriented but isotropic. Based on those XRD patterns, the lattice constants were estimated with an accuracy of 0.002Å and are shown in Fig.1. These values of *c* are smaller by about 1.3%, except for the sample with N=200, as compared to the bulk values of a=4.290Å, c=6.126Å.[12]

The in-plane SEM images of the films for N=10, 40 and 200 are shown in Fig. 2(a),(c) and (e), respectively. Both the SEM and TEM images show the "*Volmer-Weber*" type island structure, in very similar to the case of the LTP MnBi films fabricated onto the single crystal MgO(100) substrate and discussed earlier.[11] Although the size of those islands varies locally from place to place, their average height and size of those islands for N=10 ~40 are a few hundred nm and a few µm, respectively. The island height is much larger than the nominal thickness for the multilayers of N=10 ~40 prior to annealing, which are 50nm and 200nm, respectively. For the sample for N=200, the elongated islands are seen with the average height of about 1.5µm, and the length of a few tens of µm. It is noted that besides those islands clearly seen, there are smaller islands observed on the SEM images. Observations by EDX reveals that within each island, Mn and Bi atoms are uniformly distributed, as shown in Fig.2(b) and (d) for N=10 and 40.

In order to examine quantitatively the coverage of the islands, a software program of "*ImageJ*" was used,[13] where the SEM and optical images were used for N = (20~40), and for N=(10 and 200), respectively. For this analysis, the contrast was classified into the three types: i) a high contrast region which correspond to high islands, ii) medium contrast corresponding to low height islands and iii) dark image corresponding to no coverage of islands. The result is shown Fig.3, where the three different types of the coverage are shown. They are (a) *Bare substrate* where no islands are present, (b) *Low islands,* the height of which is low, and (c) *High islands,* for which the height is high. It is clearly seen that the coverage by the *Low islands* increases with N drastically to about 60% at N=40

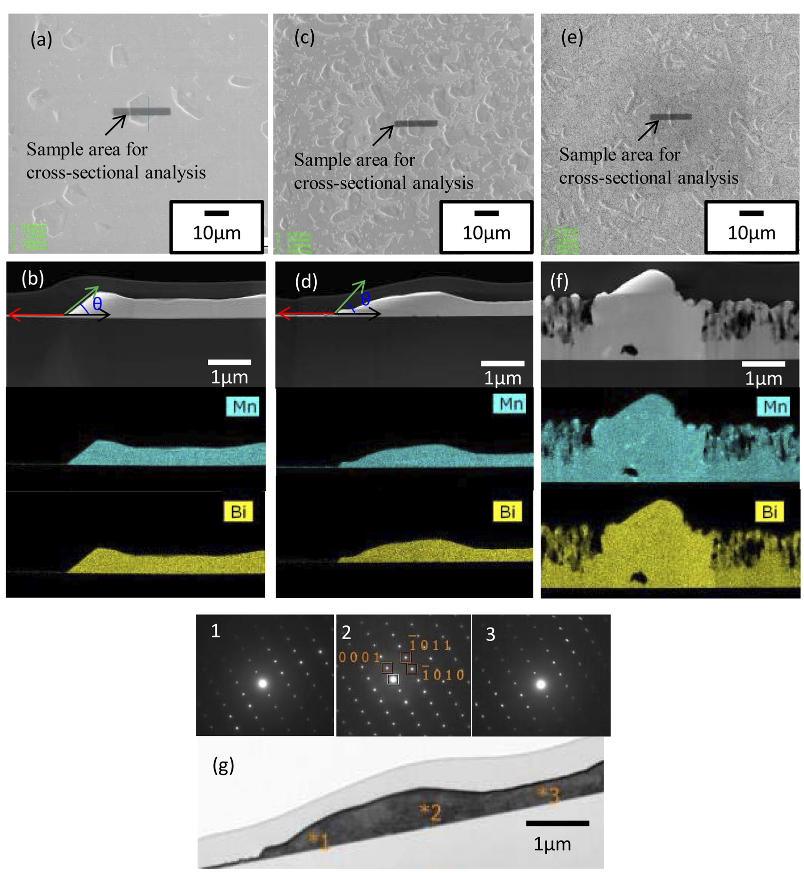

FIG. 2. In-plane SEM image and cross-sectional TEM image of the films: (a), (b) N=10. (c),(d) N=40. (e), (f) N=200. (g) electron diffraction pattern for N=40.

and 90% at N=200, while that for the *High islands* remain nearly constant (about 20%). It is also of interest to note that the average area of an island is nearly the same for N=10~40.

Figure 4 shows the out-of-plane M-H curves of the samples for N=20, 40 and 200 at various temperatures between 5 and 400K. One can see that the squareness of the M-H curves for N=20 at

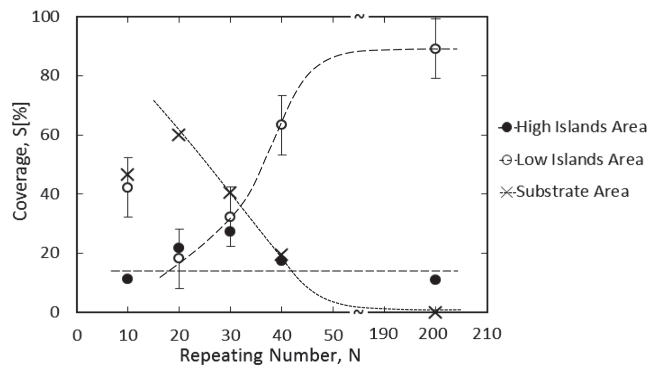

FIG. 3. The coverage S of the three types of the coverage for the island structured area as a function of the repetition number N.

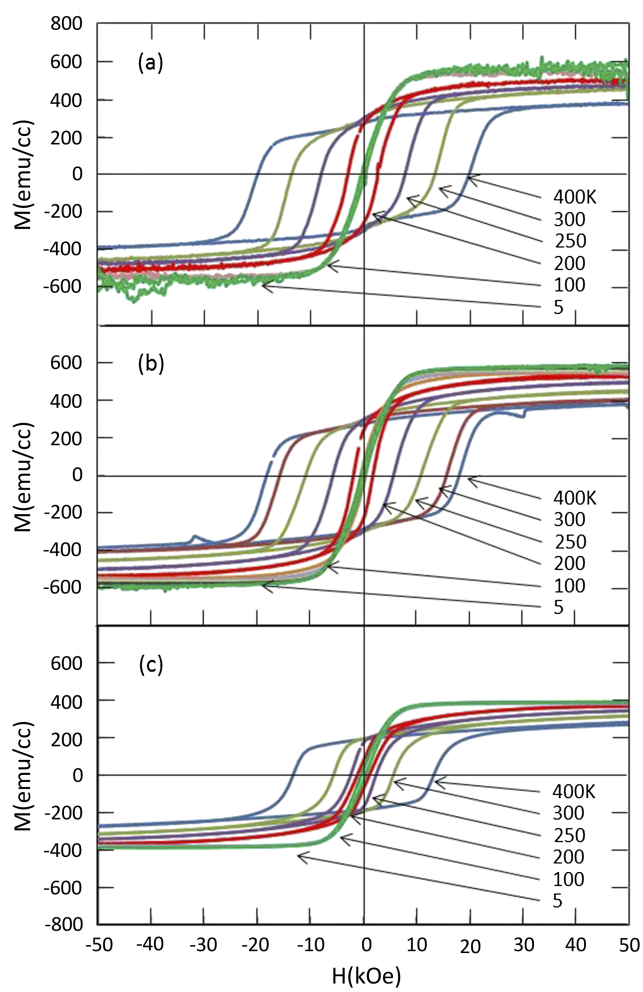

FIG. 4. The out-of-plane M-H curves of the films for (a) N=20, (b) N=40 and (c) N=200 at various temperatures.

T above 200K is high (nearly 1), whereas that for T =100K is much lower. The M-H curve at T=5K exhibits very little coercivity. The same trend is found for the sample with N=40 and 200.

Figure 5 shows (a) the saturation magnetization M_s and (b) coercivity H_c as a function of temperature for the samples with the various N values. All the temperature dependences of M_s for the various N in Fig. 5(a) are qualitatively consistent with the temperature dependence for bulk, but smaller by about 20% than those reported for bulk.[3] The reason for this discrepancy is believed to be due to the overestimation of the magnetically active volume, which was assumed to be the same as that of the nominal sample volume. According to the previous studies, the segregation of Bi and Mn were found, although they were not clearly identified by X-ray diffraction, but high resolution TEM revealed such segregations.[8] Fig. 5(b) shows the temperature dependence of H_c for the various N. The H_c is nearly zero for T below 150K, and then starts to increase with T for T above 200K. It is interesting to note that all the samples with the different N values exhibit the very similar behavior of H_c vs. T.

In order to determine the magnetic anisotropy constant Ku, measurements of the out-of–plane torque curves were carried out in fields up to 90 kOe at different temperatures from 5 to 400K. Then the torque curves were analyzed by Fourier expansions in the form of $L = A_1\sin\theta + A_2\sin2\theta + A_3\sin3\theta + A_4\sin4\theta$, where L is the torque and θ is the angle between the magnetization and the film normal. The uniaxial magnetic anisotropy constant K_u (= $K_{u1}+K_{u2}$) value is estimated based on the field dependence of the amplitude (A_2) of the sin2θ component, by extrapolating them to the infinite field. Here the demagnetization effect ($2\pi M_s^2$) was taken into account. Fig. 6(a) shows the temperature dependence of K_u thus obtained for the samples with the various N. The K_u increases monotonously with T, reaching to the values of (0.5 ∼ 1) x 10^7 erg/cc at 300K.

The second magnetic anisotropy constant K_{u2} was also estimated based on the amplitude (A_4) of sin4θ component measured at various fields, and by extrapolating them to the infinite field strength. The value of K_{u1} was then obtained as $K_{u(H\to\infty)}$ - $K_{u2(H\to\infty)}$. The result is shown in Fig. 6(b) and (c). The K_{u1} is found to increase with T, reaching to about 1 x10^7 erg/cc at 300K. On the other hand, the K_{u2} remains nearly zero for T below 300K, and increases in negative with T beyond about 250 K. At around 400 K, the K_{u2} becomes -0.8 x 10^7erg/cc. It is noted that to the best of the authors' knowledge, this is the first to report on the temperature dependence of K_{u1} and K_{u2} for LTP MnBi. It is also noted

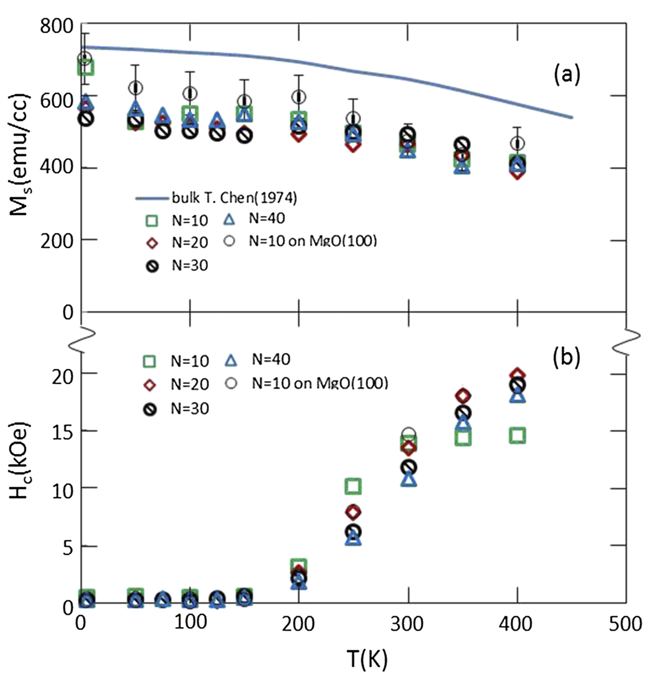

FIG. 5. The temperature dependence of (a) the saturation magnetization M_s, together with the bulk data[3] and (b) coecivity H_c of the films for the various N.

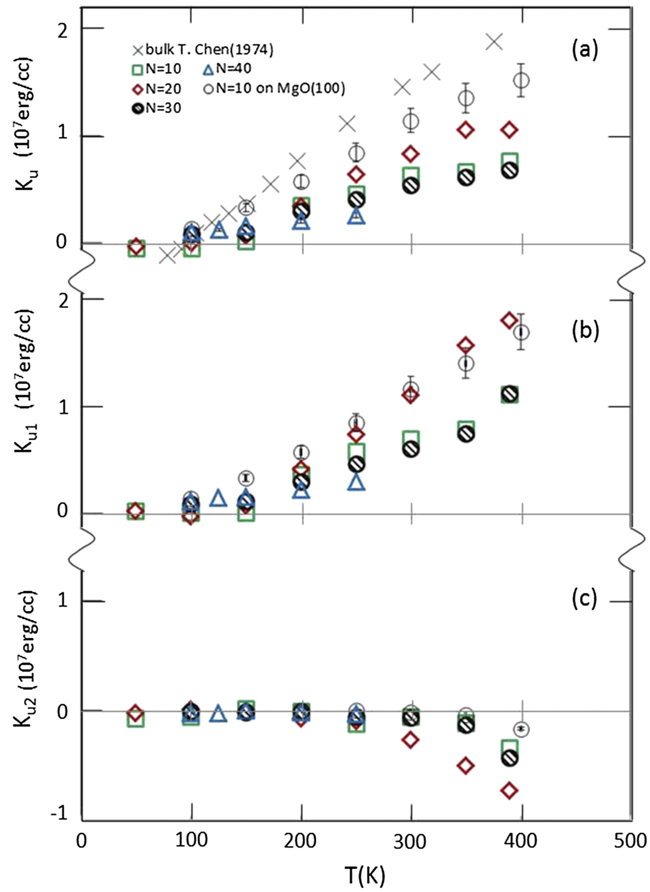

FIG. 6. The temperature dependence of the magnetic anisotropy constant (a) K_u, (b) K_{u1}, and (c) K_{u2}.

that the decrease of K_u for a temperature range beyond around 450 K is possibly due to the decrease of the Ku_2 component, as demonstrated in the present study.

IV. SUMMARY

The magnetic properties of the low temperature phase (LTP) MnBi thin films of islands structure are discussed. The LTP MnBi islands are formed onto silica substrates after the multilayers {Bi(3.2nm)/Mn(2nm)}x N are deposited and then annealed at 450°C for 0.5hr, where N is the number of the repetition of a pair of Mn and Bi layer. Those islands are found to be of the LTP MnBi, with the c-axis orientation along the normal to the sample plane for N=10 ~ 40. Their size vary from place to place, but are averagely of about a few hundred nm in height and a few µm in width for N from 10 to 40. For N=200, the elongated islands are formed densely, with the length of about a few tens of µm. The coverage of those islands increases with N. The temperature dependence of saturation magnetization M_s is qualitatively similar to that for bulk, though the absolute values for M_s are smaller by 20%. The magnetic anisotropy constants of K_{u1} and K_{u2} are evaluated for the samples with N=10 ~ 40, where K_{u1} and K_{u2} are the magnetic anisotropy constants corresponding to the second and fourth power term in the uniaxial magnetic anisotropy energy expression. It is found that the K_{u1} increases with T monotonously, reaching to about $1\text{x}10^7$ erg/cc at 400K. On the other hand, the K_{u2} remains nearly zero for temperatures below 300K, and then becomes negative, reaching to about 7 x 10^6 erg/cc at 400K. This is the first to report of the temperature dependence of K_{u1} and K_{u2} in the LTP MnBi of an island structure. It is also noted that the decrease of K_u for a temperature range beyond around 450 K is possibly due to the decrease of the Ku_2 component, as demonstrated in the present study.

ACKNOWLEDGMENTS

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