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Structural, magnetic and magneto-transport properties of Pt-alloyed MnBi thin films

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The structural, magnetic and magneto-transport properties of highly c-axis oriented Mn$_{55-x}$Pt$_x$Bi$_{45}$ ($x=0$, 1.5, 3, and 4.5) thin films have been investigated. The coercivity of the Pt-alloyed thin films increases and the saturation magnetization decreases as the Pt concentration increases. The anisotropy field $H_a$ increases as a function of Pt concentration, too but the coercivity increases more rapidly than the anisotropy field. This indicates an enhanced domain-wall pinning, caused by increased interstitial disorder due to the occupancy of regular Mn sites by Pt. The same mechanism explains the reduced magnetization. All samples exhibit a large extraordinary Hall effect with anomalous Hall coefficient about an order of magnitude larger than the ordinary Hall coefficient.


I. INTRODUCTION

There has been considerable interest in understanding the magnetic, optical, and magneto-transport properties of MnBi-based materials because of their substantial magneto-optical and permanent magnet properties. MnBi exhibits interesting properties including an extraordinarily large Kerr rotation, a Curie temperature well above room temperature, a high coercivity with a rectangular hysteresis loop, and a large perpendicular anisotropy in thin films at room temperature.

Pure MnBi adopts the hexagonal NiAs-type crystal structure at room temperature with lattice constants of $a=4.29$ Å and $c=6.12$ Å. Around 628 K, it undergoes coupled structural and magnetic phase transitions from a ferromagnetic low-temperature phase (LTP) to a paramagnetic high-temperature phase (HTP). If MnBi in the HTP is rapidly quenched, a ferromagnetic quenched HTP (QHTP) with a Curie temperature of about 440 K is obtained. QHTP is thermally unstable and transforms slowly back to LTP at room temperature with a time constant of about two years. In addition to the structural instability, MnBi in LTP shows an increasing coercivity as temperature increases reaching a maximum at 553 K. These circumstances have led to several studies of the effect of dopants on the various properties of this material. Some of these experiments show that the structural, magnetic, and magneto-optical properties of MnBi can be controlled by doping it with a third element. The goal of this study is to understand and control the structural, magnetic, and magneto-transport properties of MnBi-based alloys by alloying with the heavy and noble element Pt.

II. EXPERIMENTAL METHODS

We have synthesized Pt-alloyed MnBi thin films having post-annealed stoichiometry of Mn$_{55-x}$Pt$_x$Bi$_{45}$ ($x=0$, 1.5, 3, and 4.5) by sequential evaporation of Bi, Mn, and Pt onto a glass substrate at room temperature using an AJA e-beam evaporation system. The base pressure of the evaporation chamber was $4 \times 10^{-9}$ Torr. Bi and Mn were evaporated from alumina crucibles and Pt was evaporated from a graphite crucible. The atomic ratios of the constituent elements in the Mn$_{55-x}$Pt$_x$Bi$_{45}$ films were estimated from the respective layer thicknesses and were later confirmed using energy dispersive x-ray (EDX) spectroscopy. The deposition layer thickness was controlled by a quartz crystal thickness monitor. All the samples were annealed in situ immediately after deposition at a temperature of 375 °C for one hour. The thicknesses of the postannealed samples were determined from x-ray reflection measurement using a Bruker X-ray diffractometer, and for the structural characterization of the thin films a Rigaku X-ray diffractometer was used. Magnetic properties were investigated using Quantum Design magnetic property measurement system (MPMS) and magneto-transport properties were studied using Keithley electrometers in conjunction with MPMS.

III. RESULTS AND DISCUSSION

Figure 1 shows the x-ray diffraction (XRD) patterns of Mn$_{55-x}$Pt$_x$Bi$_{45}$ ($x=0$ and 4.5) thin films collected at room
samples. We have found that the saturation magnetizations of Mn55−xPt4.5Bi45 thin films. $M_s(H)$ and the coercivity $H_c$ of the samples with Pt concentration. Inset (left): Temperature dependence of the magnetization for Mn50.5Pt4.5Bi45 thin film measured in two perpendicular directions.

FIG. 2. (Color online) Magnetization as a function of magnetic field for Mn55−xPt4.5Bi45 thin films. Inset (right): Change in the saturation magnetization $M_s$ and the coercivity $H_c$ of the samples with Pt concentration. Inset (left): Temperature dependence of the magnetization for Mn50.5Pt4.5Bi45 thin film measured in two perpendicular directions.

Thin film quality can also be assessed from the nature of the top and bottom interfaces of the thin film. Pt substitution has increased the roughness of the interfaces. The faster damping of x-ray reflectivity oscillations in the Pt alloyed samples, as shown in the inset (i) of Fig. 1, is a consequence of the rough interfaces. The change in the surface morphology due to Pt substitution can be clearly seen in the atomic force microscope micrographs as well. The surface of Mn50.5Pt4.5Bi45 film is continuous with root-mean-square roughness of 1.7 nm over a 4 × 4 μm² scan area and that of Mn55Bi45 film decreases from 7.03 × 10⁶ ergs/cm³ for Mn55Bi45 thin films to 5.60 × 10⁶ ergs/cm³ for Mn50.5Pt4.5Bi45 films.

As explained in the introduction, the phase structure of MnBi is very complicated, even in the bulk. However, a key feature of the system is the presence of large bipyramidal interstices in the NiAs structure, which can easily be occupied by the Mn atoms. As pointed out by Roberts, each of these interstitial atoms is coordinated by five Mn atoms so that a single Mn atom with antiferromagnetic coupling yields a substantial reduction in the moment. Such a reduction is very likely because Mn and most Mn compounds exhibit a pronounced trend toward antiferromagnetism (AFM). AFM is generally favored for half-filled shells because the energy of electron in a filled 3d↑ subband is equal to the on-site energy, whereas the antiferromagnetic state can reduce its energy by interatomic hopping. Ordered MnBi is an exception, with a atomic structure and a charge state (essentially Mn³⁺) supporting ferromagnetism. In the present system, the Mn-rich regions around the interstices are closer to antifer-
romagnetic bulk Mn. The excess Mn pushes the system toward antiferromagnetic (or noncollinear) order, and the Pt atoms further increase the interstitial Mn occupancy by displacing the Mn atoms from their regular sites. The effect is further enhanced by the Pt atoms on the regular lattice sites, which further dilute and probably reverse the FM interactions of MnBi.

The coercivity scales as $H_c = 2K_1/M_s$, but it also exhibits a strong dependence on the defect structure and some dependence on $K_2$. Since $K_2$ does not change very much, from $2.5 \times 10^6$ ergs/cm$^3$ for Mn$_{55}$Bi$_{45}$ to 2.06 $\times 10^6$ ergs/cm$^3$ for the sample with 4.5% Pt, we ignore the effect of $K_2$ on the coercivity. The $K_1$-only anisotropy field increases from $H_{K_1} = 25.4$ kOe (0% Pt) to 34.5 kOe (4.5% Pt) but this increase is insufficient to explain the observed increase in coercivity. The ratio $H_c/H_o$ increases from 0.185 for 0% Pt to 0.362 for 4.5% Pt. Both ratios, especially the second one, are large and indicate a very effective pinning mechanism. The increase due to Pt substitution indicates substantial structural inhomogeneities, which is consistent with our finding that the Pt enhances deviations from the perfect NiAs structures and also produces discontinuities on the film surface. Details of the pinning mechanisms are not known at present but the domain wall energy $\gamma = 4AK_1^{1/2}$ strongly depends on the interatomic exchange $\Delta = z J$, so that nanoscale regions with strongly reduced exchange $J$ and/or $z$ are effective pinning centers.

Besides investigating structural and magnetic properties, we have studied the galvanomagnetic properties of Mn$_{55-}^{55-}Pt_{Bi_{45}}$ thin films. We have carried out Hall measurement on these samples using the van der Pauw method. The samples exhibit a large extraordinary Hall effect. Hall resistivity $\rho_{xy}(H)$ loops for samples with platinum concentration of 0, 1.5, and 2% are essentially identical to their Mn(H) hysteresis loops, as shown in Fig. 4. However, $\rho_{xy}(H)$ for the sample with 4.5 at. % Pt shows a quadratic dependence on magnetic field, indicating that the ordinary magnetoresistance starts dominating as the Pt concentration increases, as shown in the left inset in Fig. 4. We have determined the normal and anomalous Hall coefficients $R_o$ and $R_y$ by fitting

$$\rho_{xy} = R_o B + 4\pi R_y M_s$$

for the high field $\rho_{xy}(H)$ data, where B is the magnetic induction and $M_s$ is the saturation magnetization. For all the samples, the $R_y$ values are about one order of magnitude larger than $R_o$ values, which is consistent with past research. As shown in the right inset of Fig. 4, $R_y$ increases from $1.26 \times 10^{-10}$ to 1.49 $\times 10^{-10}$ $\Omega$ cm/G and $R_o$ decreases from $5.7 \times 10^{-12}$ to 1.0 $\times 10^{-13}$ $\Omega$ cm/G as the Pt concentration increases from 0 to 3 at. %.

We could not precisely determine the values of $R_o$ and $R_y$ for the sample with 4.5% Pt due to the quadratic dependence of $\rho_{xy}$ on H. The increase in $R_y$ and decrease in $R_o$ is consistent with the increase in resistivity of the samples due to Pt substitution.

IV. CONCLUSIONS

We have investigated the structural, magnetic and magneto-transport properties of Mn$_{55-}^{55-}Pt_{Bi_{45}}$ (x=0, 1.5, 3, and 4.5) thin films prepared by e-beam multilayer deposition and annealing. We have found that the magnetic properties of MnBi are very sensitive to the substitution of a third element such as Pt. Saturation magnetization and coercivity of the samples show a systematic change with the Pt concentration. The substantial decrease in magnetization and increase in the coercivity of the Pt-alloyed samples can be attributed to the defect structure with strong pinning centers. The increase in the resistivity of the samples due to platinum substitution causes an increase in the extraordinary Hall coefficient and decrease in the ordinary Hall coefficient.

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19The nucleation-field coercivity is independent of $K_2$ but depending on the real structure, this is not necessarily true for pinning.