Magnetic anisotropy properties and spin reorientation for textured Bi-Mn alloys fabricated by a field-inducing technique

Yongsheng Liu, Jincang Zhang,* Guangqiang Jia, Xiaoyong Zhang, Zhongming Ren, Xi Li, Chao Jing, Shixun Cao, and Kang Deng

Department of Physics and Department of Materials Science & Engineering, Shanghai University, Shanghai 200436, People's Republic of China

(Received 22 April 2004; revised manuscript received 12 July 2004; published 17 November 2004)

By a magnetic-field-inducing technique, Bi-Mn alloys were fabricated with the textured structure and anisotropic characteristics. Magnetic properties of MnBi compound aligned in alloys with 6 wt % Mn have been investigated systematically. The saturation magnetization M_s decreases with the increase of temperature. At temperatures below 150 K, the coercive field H_c decreases with the increase of temperature, while the coercive field H_c increases with temperature above 150 K. In the range of temperature from 150 to 300 K, the remanent magnetization M_r and the M_r/M_s increase with the temperature, while below 150 K, the M_r and the M_r/M_s reach a constant value of nearly zero. The magnetic moments rotated from being parallel to the *c* axis toward the basal plane for MnBi (the low-temperature phase) at about 90 K. Most of all, under a dc magnetic field applied parallel to the *c* axis, the transition temperature of the spin reorientation decreases with the increase of magnetic field, and even decreases 30 K under 5 T. The mechanism of the spin-reorientation transition and the change of its transition temperature are discussed and explained by phenomenological theory.

DOI: 10.1103/PhysRevB.70.184424 PACS number(s): 75.30.−m, 75.25.+z, 75.47.Np, 75.30.Kz

I. INTRODUCTION

As one of the most powerful tools available to researchers for the study, modification, and control of matter, the high magnetic field has caused many interesting phenomena[1–6] and is used in various academic fields such as materials science and physics.[7–12] Currently, the physics of high-field-induced transition[13–19] is among the most interesting problems in condensed matter physics. Owing to the high uniaxial magnetic anisotropy of its low-temperature phase (LTP) and the good magneto-optical properties of its quenched high-temperature phase, the physical properties of the binary compound MnBi have been investigated extensively.[20–23,26–28] MnBi (LTP) crystallizes in a hexagonal unit cell with space group $R\bar{3}m$ and lattice constants $a=4.2827$ Å and $c=6.1103$ Å.[22] The crystal structure of MnBi (LTP) is shown in Fig. 1. At 633 K, a ferromagnetic LTP with a NiAs-type structure transforms to a paramagnetic high-temperature phase (HTP) with a Ni_2In-type structure on heating. However, the magnetic transition temperature is 600 K when cooling.[23] MnBi compound has considerable potential as a hard magnet at high temperatures and as a permanent phase in nanocomposite magnets because the coercivity of the LTP increases with temperature.[22] Moreover, MnBi film is a good candidate for magneto-optical materials because of its high Kerr rotation.[20] In order to investigate the effect of magnetic field on the structure and magnetic properties of MnBi compound, in this work, alloys with 6 wt % Mn (Bi-6 wt % Mn) with textured structure and anisotropy characteristics were prepared by a magnetic-field-inducing technique. Magnetic properties of MnBi compound aligned in the alloys have been investigated systematically. The results reveal an existence of spin-reorientation transition (SRT) at approximately 90 K in MnBi (LTP). During SRT the magnetic moments rotated from being parallel to the *c* axis toward the basal plane, and the spin-orbit interaction plays a dominant role in the anisotropy of MnBi (LTP). It is noticed that different measurements of dc magnetic fields H_m along the *c* axis result in different temperatures of SRT.

II. EXPERIMENTAL PROCEDURE

Since the investigation of MnBi compound exhibits an easy axis of magnetic anisotropy below about 630 K, a magnetic alignment method was carried out by means of solidification in different magnetic fields H_f. Details of the experimental technique were described in previous papers.[24,25] Bi-Mn alloys with composition of 6 wt % Mn were prepared using bismuth (99.0% purity) and electrolytic manganese (99.5% purity). The alloys were melted in an inductive furnace and cast to graphite molds under argon at a pressure of 50.6 kPa. The samples, 9.5 mm in diameter and 25 mm in length, were sealed in a graphite tube and inserted into a resistance furnace placed between two poles of the electromagnet, as shown in Fig. 2. The intensity of the magnetic field between poles of the electromagnet can be adjusted and the temperature in the furnace chamber can be controlled automatically during the experiment. Since the liquidus temperature of Bi-6 wt % Mn alloy is above 630 K, the alloy is in a semisolid state at 548 K. The alloys were heated up at a rate of 10 K/min to the temperature 548 K in mushy zone without magnetic field, then held for 30 min and cooled to the temperature below 535 K under a dc magnetic field ($\boldsymbol{H}_f$) of 0.5 T. The cooling rate was about 0.15 K/s. For characterizing the morphology of the MnBi phase, the samples obtained in the experiments were mechanically polished parallel and perpendicular to the $\boldsymbol{H}_f$ direction, respectively. It is easy to understand that the cylinder axis of aligned MnBi phase, being identical to the $\boldsymbol{H}_f$ direction during the magnetic-solidification procedure, is the c-axis direction of

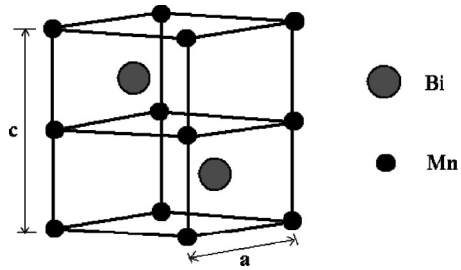

FIG. 1. Crystal structure of MnBi (LTP).

MnBi (LTP); i.e., the easy magnetization direction. The structure of the samples was characterized by optical microscopy, scanning electron microscopy, and x-ray diffraction (XRD) with Cu-K_α radiation, respectively. Differential thermal analyzer (DTA) was used to study the phase transitions at high temperature, and magnetization measurement was conducted to investigate the magnetic properties and phase transitions at low temperature. Magnetic measurements were performed under the applied magnetic field $\boldsymbol{H}_m$ up to 50 kOe in the temperature range from 1.9 to 300 K using a physical properties measurement system (PPMS/Quantum Design Inc.). From the principle, during dc measurement a constant field is applied and the sample is moved quickly through both an ac-drive coil set and a detection coil set, inducing a signal in them according to Faraday's law. This measurement method is commonly called the extraction method.

III. RESULTS AND DISCUSSION

Figure 3 shows the microstructures of the Bi-6 wt % Mn alloy solidified under a dc magnetic field $\boldsymbol{H}_f$=0.5 T. In the case of solidification in the magnetic field, the elongated MnBi crystals (dark gray) are oriented and grow preferentially along the direction of field in the Bi matrix (white) [Fig. 3(b)]. The hexagonal sections of the crystals only appear in the section perpendicular to the field [Fig. 3(a)]. Furthermore, XRD patterns show that the *c* axis of hexagonal MnBi crystal (easy magnetization axis) is aligned parallel to the $\boldsymbol{H}_f$ direction. By comparison with magnetic powders aligned under magnetic field in an epoxy resin to form a bonded magnet,[26] the present method has a prominent feature that MnBi (LTP) crystals not only align along the *c* axis, but also grow preferentially and congregate along the $\boldsymbol{H}_f$ direction. Only peaks of Bi and MnBi (LTP) are observed and no peak of Mn appears in the XRD patterns of the solidified Bi-6 wt % Mn alloy. The XRD results and energy dispersive X-ray analysis indicate the formation of MnBi (LTP) (approximately 28.656 wt %) and Bi phase. Because Bi is diamagnetic, its effect has been ignored and only MnBi (LTP) has been considered in the magnetization measure-

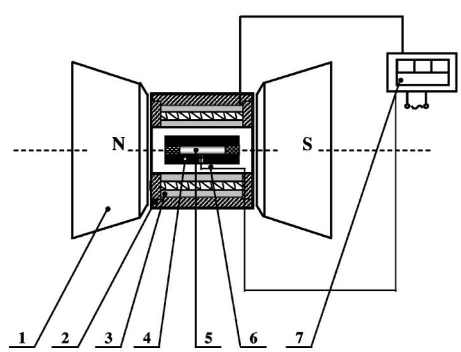

FIG. 2. Schematic diagram of the experimental device of metal solidification in magnetic field. 1—Electromagnet yoke, 2—Refractory, 3—Heater, 4—Graphite tube, 5—Sample, 6—Thermocouple, 7—Temperature controller.

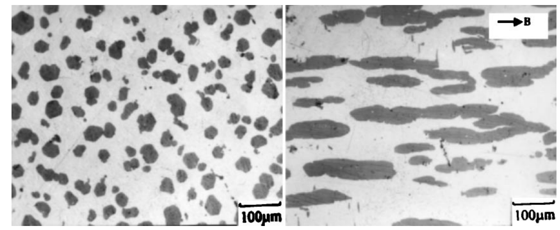

FIG. 3. Microstructures of Bi-6 wt % Mn alloy solidified in a dc magnetic field $\boldsymbol{H}_f$=0.5 T: (a) section perpendicular to $\boldsymbol{H}_f$; (b) section parallel to $\boldsymbol{H}_f$.

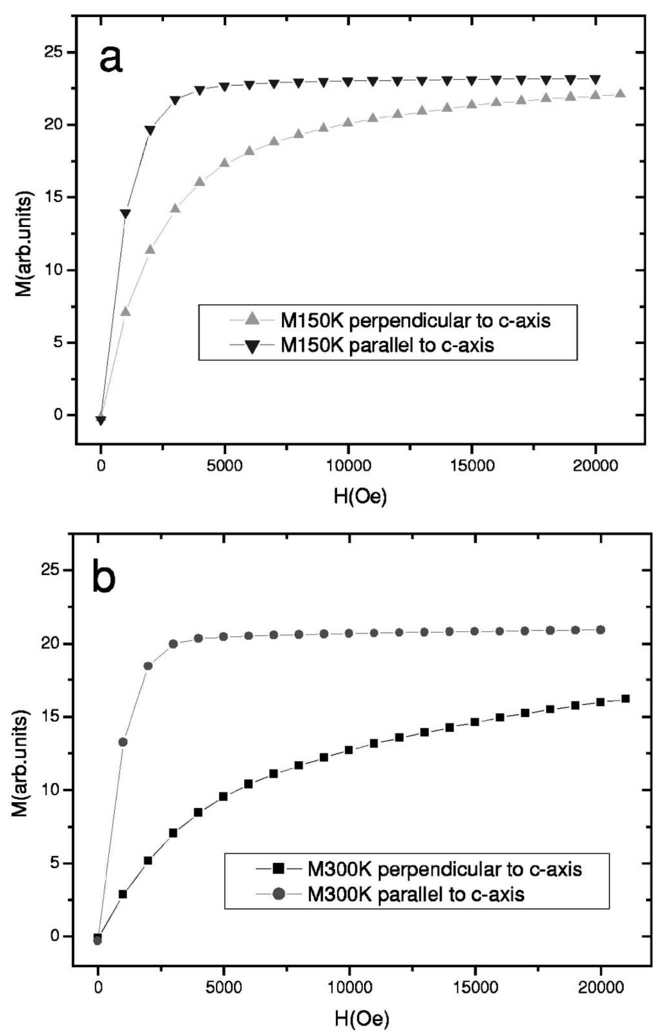

FIG. 4. Magnetizations parallel and perpendicular to the *c*-axis direction at 150 and 300 K.

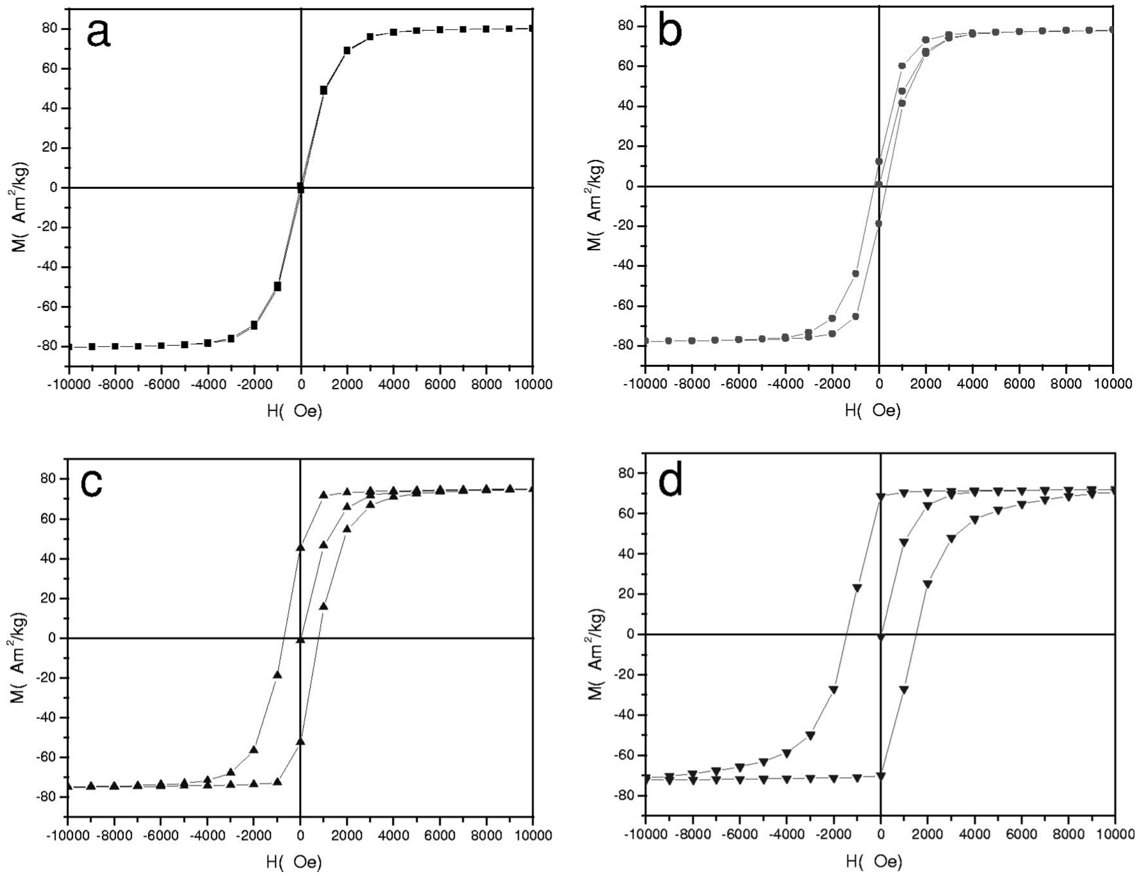

FIG. 5. Hysteresis loops of MnBi (LTP) compound measured along the c axis at 150, 200, 250, and 300 K.

ment. The MnBi crystals observed in the microstructure micrographs are randomly oriented in the Bi matrix in the sample crystallized without the magnetic field. Results of DTA showed that there are phase transitions at about 544 and 630 K, corresponding to the Bi melting and the transformation of the ferromagnetic LTP to the paramagnetic phase in MnBi, respectively.

To study the magnetic anisotropy, magnetization curves along and perpendicular to the c-axis direction were measured at different temperatures. Figure 4 presents the magnetizations parallel and perpendicular to the c-axis direction at 150 and 300 K. It can be seen that when the applied field H_m is parallel to the c axis, saturation occurs much more easily compared with the case H_m perpendicular to the c axis. The anisotropy fields in MnBi (LTP) at 150 and 300 K are about 2.5 and 5 T, respectively. This suggests that there is a very strong anisotropy in the MnBi compound and the anisotropy field of MnBi (LTP) increases with the increasing temperature. In Fig. 5, hysteresis loops of MnBi (LTP) compound measured along the c axis at 150, 200, 250, and 300 K are displayed, respectively. It is easy to see that the saturation magnetization M_s decreases, but the coercive field H_c and the remanent magnetization M_r increase with the increase of temperature. The saturation magnetization M_s and the coercive field H_c of MnBi alloy along the c axis at various temperatures are shown in Fig. 6. It is clear that M_s decreases with the increase of temperature, and H_c decreases with the increase of temperature below 150 K, but increases above 150 K. M_s shows spin-wave behavior and holds fairly well the $T^{3/2}$ law, especially at low temperature. In the single crystal MnBi (LTP), Chen and Stutius found that the uniaxial anisotropy energy increases from -2.5×10^5 J/m^3 at 4.2 K to 3×10^5 J/m^3 at 150 K, while the in-plane anisotropy energy decreases from 16×10^5 J/m^3 to 4×10^3 J/m^3; the uniaxial anisotropy energy goes up further with increasing temperature and reaches a maximum of 2.2×10^6 J/m^3 at 490 K.[23] Hence, the increase of coercivity with the increase of temperature can be attributed to the increase of anisotropy energy in MnBi (LTP). The inset in Fig. 6 is the remanent magnetization M_r and the M_r/M_s as functions of temperature. The tendencies of the remanent magnetization M_r and M_r/M_s are same. For $150\text{ K}<T<300\text{ K}$, M_r and M_r/M_s increase with temperature, while below 150 K M_r and M_r/M_s reach a low saturation value, nearly zero. For a ferromagnetic single crystal with uniaxial anisotropy, $M_r = M_s \cos x$, x is an angle between the applied field and the easy magnetization axis.[29] Therefore, it can be concluded that below 150 K, the easy axis is in, or nearly in, the basal plane for MnBi (LTP).

The effect of the magnetic field on the temperature of SRT can be seen from the thermomagnetic curves $M(T)$ presented in Fig. 7. The $M(T)$ curves for MnBi (LTP) compound were obtained under different magnetic fields along the c-axis direction of MnBi (LTP). In Fig. 7, under an applied dc magnetic field $\boldsymbol{H}_m$, the value of magnetization along the c axis increases slowly and the shape of the anomaly in mag-

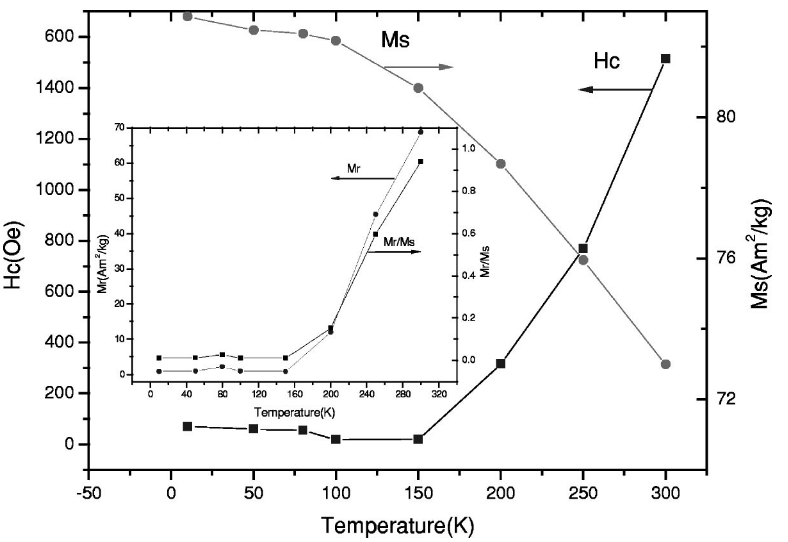

FIG. 6. Saturation magnetization M_s and the coercive field H_c of MnBi (LTP) compound along the c axis at various temperatures. The inset shows the remanent magnetization M_r and the M_r/M_s as functions of temperature.

netization for higher dc field becomes nonpeaklike (a peaklike profile indicates a spin reorientation of MnBi (LTP) for lower dc field, also see the inset in Fig. 8). The position of the peak or the bump of magnetization, corresponding to the spin reorientation temperature, shifts to lower temperature with increasing $\boldsymbol{H}_m$ (Figs. 7 and 8). The temperature of SRT as a function of magnetic field $\boldsymbol{H}_m$ applied along the c-axis is plotted in Fig. 8. It shows that the temperature of SRT decreases with the increase of applied field. This fact means that although the Zeeman energy $(-\boldsymbol{M}\cdot\boldsymbol{H})$ produced by the applied dc field keeps the magnetization parallel to the field direction, the external dc field modifies the temperature of SRT: T_s even decreases 30 K when the dc magnetic field 5 T is applied parallel to the c-axis direction. This also implies that the anisotropy energy is comparable to the Zeeman energy produced by a certain applied magnetic field. In order to further study the influence of an applied magnetic field on the temperature of SRT, the free energy in the c-axis direction and the basal plane at various temperatures should be calculated, which is underway.

Generally, the magnetic moments rotated from along the c axis to, or nearly to, the basal plane for MnBi (LTP) at about 90 K, which has been observed in many previous experiments.[23,28,30,31] The first self-consistent spin-polarized band-structure calculation for MnBi (LTP) indicates that the average local magnetic moment on the Mn atoms is 3.6 μ_B and *pd* hybridization induces a negative magnetic moment of -0.08 μ_B on the Bi site.[32] The calculation results of Yang *et al.* also show that the Mn atom possesses a magnetic moment of 3.6 μ_B, and that the Bi atom has a magnetic moment of -0.15 μ_B due to the *sd* and *pd* hybridization between Bi and Mn atoms.[26] Since the moments of the Mn-lattice and Bi-lattice ions are aligned antiparallel to each other along the easy c axis, it is perhaps expected that, similar to the spin flipping observed in ferrimagnetic materials, the interstitial Bi ions may reverse their orientation at some critical field to reduce the total energy. However, the critical field is expected to be much larger if the spin reversal in MnBi (LTP) is similar to the spin-flipping in ferromagnetic materials, of which the critical field is several hundred MA/m (several MOe).[33] Therefore, it can be concluded that the anomalous

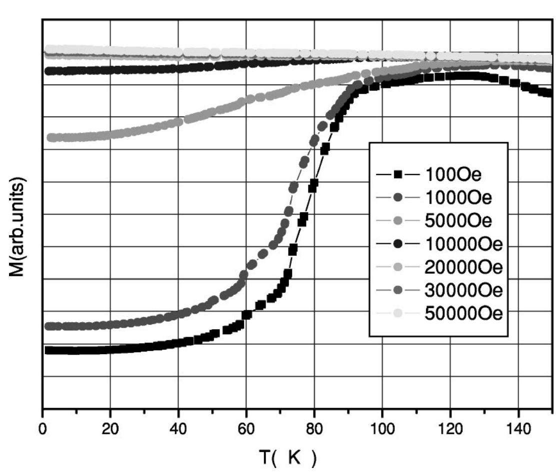

FIG. 7. Temperature dependence of the magnetization measured on MnBi (LTP) compound when different magnetic fields ($\boldsymbol{H}_m$) are applied along the c-axis direction.

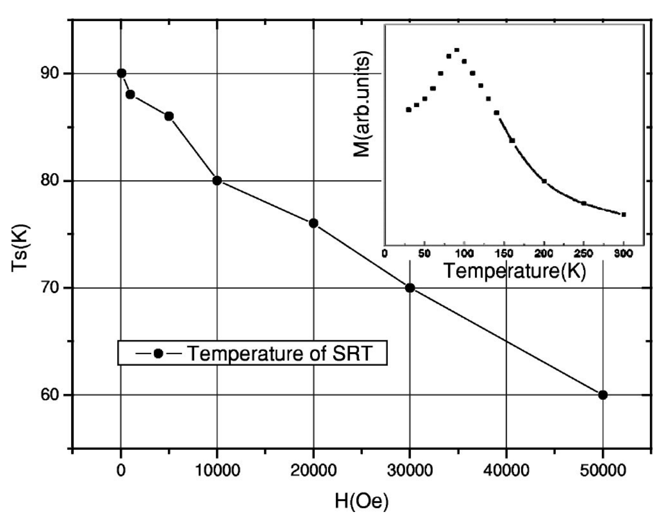

FIG. 8. Temperature of SRT as a function of magnetic field $\boldsymbol{H}_m$ applied along the c axis. The inset shows the temperature dependence of magnetization ($\boldsymbol{H}_m$=2000 Oe) for MnBi along the c axis by Ref. 22.

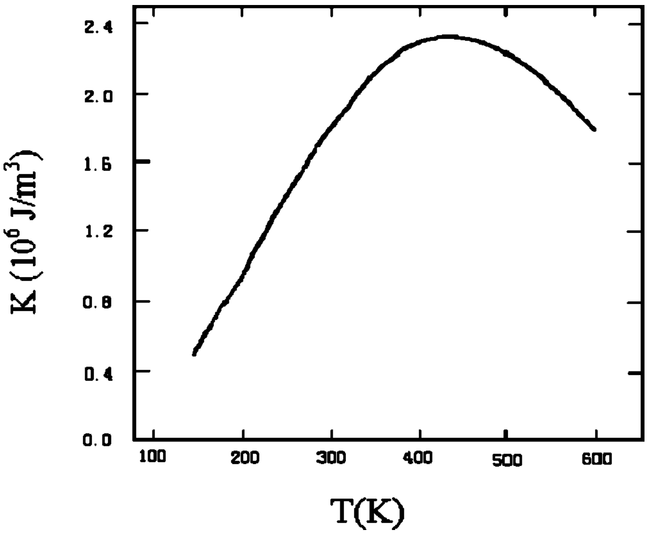

FIG. 9. Anisotropy constant $K=K_{u1}+2K_{u2}+3K_{u3}$ as a function of temperature (Ref. 34).

behavior in $M(T)$ does not originate from the spin reversal of MnBi (LTP).

MnBi (LTP) compound is a hexagonal crystal and the hexagonal axis is the direction of easy magnetization at room temperature. In MnBi (LTP) the anisotropy energy density is given by (ignoring the small higher-order term)

$$U_A = K_{u1}\sin^2\theta + K_{u2}\sin^4\theta, \quad (1)$$

where θ is the angle between the magnetization and the hexagonal axis; K_{u1} and K_{u2} are the first and second anisotropy constants, respectively. If $K_{u1}>0$ and $K_{u1}+K_{u2}>0$, the magnetization is stable along the *c* axis. If $-K_{u2}>K_{u1}\geqslant 0$, the anisotropy energy is maximum in the *c* axis, so that the magnetization is stable when it lies in any direction in the basal plane (perpendicular to the *c* axis, θ=90°). If $2K_{u2}>-K_{u1}>0$, the stable direction forms a cone of easy magnetization.[29] By linear extrapolation of the anisotropy constant $K=K_{u1}+2K_{u2}+3K_{u3}$ as a function of temperature (shown in Fig. 9)[34] to a temperature below 90 K, $K<0$ will probably appear, then K_{u1} and K_{u2} may meet the conditions of the plane anisotropy or the easy cone. Albert and Carr measured the anisotropy constants K_{u1}, K_{u2}, and K_{u3} in oriented polycrystals of MnBi at various temperatures from 77 to 300 K.[31] As a result, at 77 K, $2K_{u2}>-K_{u1}>0$ can be found, so the stable direction forms a cone of easy magnetization. This is consistent with the suggestion of Yang *et al.*[26] and Roberts.[30] Yang *et al.* found that below 200 K, the magnetic moments of Mn deviate gradually from the direction parallel to the *c* axis and turn to the direction nearly perpendicular to the *c* axis below 50 K. However, the magnetic moment still shows a very small *c*-axis component at 10 K; i.e., a conic magnetic structure is formed in MnBi from 240 to 10 K.[22] Roberts also pointed out that the magnetic moments do not rotate completely from along the *c* axis to the basal plane at temperatures below 84 K.[30] However, this does not agree with other researches that concluded that the magnetic moments flip into the basal plane at about 90 K.[23,28] For a conic magnetic structure, the angle θ between the magnetization and the hexagonal axis is $\sin^{-1}(\sqrt{-K_{u1}/2K_{u2}})$. To study the change of θ with the temperature, it is necessary to accurately measure K_{u1} and K_{u2} at low temperature.

Once a dc magnetic field has been applied along the *c* axis to the MnBi (LTP), the anisotropy energy density is given by

$$U_A = U_K + E_Z = K_{u1}\sin^2\theta + K_{u2}\sin^4\theta - M\cdot H, \quad (2)$$

where E_Z is the Zeeman term, $\boldsymbol{H}$ and $\boldsymbol{M}$ denote the external field and saturation magnetization, respectively. For fields applied in the *c*-axis direction, the Zeeman energy favors the arrangement of spins along the *c* axis. This competes with the plane anisotropy, causing a continuous rotation of spins to the *c* axis with the increase of field. Finally, with field applied along the *c* axis, a spin-reorientation transition occurs due to the competition between Zeeman energy and the plane anisotropy, resulting in a slow spin rotation to the basal plane.

A magnetic orbital moment coupled with the spin leads to a variation in the exchange or electrostatic energy in magnetization process, through a change in the overlap of wave functions. That is, the magnetization of the crystal sees the lattice through orbital overlap of the electrons: spin interacts with orbital motion by means of the spin-orbit coupling. With the decrease of temperature, the lattice parameter decreases and it results in an increase of the coupling. Generally speaking, magnetic anisotropy is produced through the interaction between spontaneous magnetization and the crystal lattice, so that the temperature dependence of spontaneous magnetization should give rise to a change in magnetic anisotropy.[33] Meanwhile, the temperature dependence of the anisotropy is stronger than that of the spontaneous magnetization, as their relationship is given by

$$K_u/K_{u0} = (M_s/M_{s0})^3, \quad (3)$$

where K_u is the anisotropy constant for uniaxial anisotropy, K_{u0} is the anisotropy constant at 0 K, M_s is the spontaneous magnetization, and M_{s0} is the spontaneous magnetization at 0 K.[35] The magnetic moment of MnBi gradually deviates from the *c* axis below about 150 K, and flips into the plane perpendicular to the *c* axis or an easy cone at approximately 90 K. The low-temperature magnetic anisotropy cannot be explained by the magnetic dipole-dipole interactions, as this would favor the *c* axis as quantization axis, the spin-orbit interaction thus plays a key role in the anisotropy at low temperature. Band structure at Γ (the center of the first Brillouin zone) as a function of the spin-orbit parameter λ displays the spin-flip effect.[32] Hence, the effect of magnetic field applied along the *c* axis on the temperature of SRT probably originates from the interaction between the magnetic field and spin-orbit coupling.

IV. CONCLUSION

By a magnetic-field-inducing technique, Bi-Mn alloys with textured structure were fabricated. The anisotropy magnetic properties and spin-reorientation transition in the MnBi compound aligned in the Bi-6 wt % Mn alloy have been studied systematically. The saturation magnetization M_s decreases with the increase of temperature, and the coercivity H_c decreases with the increase of temperature below 150 K but increases with temperature above 150 K. In the temperature range of 150 to 300 K, the remnant magnetization M_r and the ratio of M_r/M_s increases with temperature, while below 150 K, M_r and the M_r/M_s reach a constant value, nearly zero. Owing to the variations of the anisotropy constants K_{u1} and K_{u2}, the magnetic moment of MnBi gradually deviates from the *c* axis below about 150 K, and flips into the basal plane perpendicular to the *c* axis or an easy cone at approximately 90 K. It is interesting that, under a dc field H_m applied parallel to the *c* axis, the temperature of SRT decreases with the increase of magnetic field. The spin-orbit interaction plays a dominant role in the anisotropy at low temperature, and the effect of magnetic field applied along the *c* axis on the SRT temperature may originate from the interaction between the magnetic field and spin-orbit coupling.

ACKNOWLEDGMENTS

This work was supported by the National Foundation of the Natural Science of P. R. China (Nos. 50234020, 10274049), the Shanghai Nano-technology Promotion Center and the Science & Technology Committee of Shanghai Municipality (No. 0352nm036). This work was also partly supported by the Dawn Project (No. 03SG35) and Science & Technology Development Foundation of the Education Committee of Shanghai Municipality (No. 02AK42) and the Shanghai Leading Academic Discipline Program.

*Corresponding author. Email address: jczhang@mail.shu.edu.cn

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