# Magnetic properties and microstructure of amorphous $Co_{100-x}Tb_x$ thin films

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Amorphous $Co_{100-x}Tb_x$ alloy thin films with the composition of $x=7-60$ at. % were prepared by dc magnetron sputtering at various powers and argon pressures then annealing in vacuum. Effects of the composition, sputtering power, argon pressure, and annealing temperature on the parallel and normal to the film plane magnetic properties have been investigated. The analysis of transmission electron microscopy diffraction patterns and magnetic measurement data indicate that an amorphous film with isotropic magnetic properties can be produced after low temperature annealing. The maximum observed perpendicular coercivity was as high as 6000 Oe for the as-deposited Co–Tb amorphous films. A nearly magnetically isotropic amorphous Co–Tb film with in-plane coercivity of about 2080 Oe was obtained after annealing. © *1998 American Institute of Physics.* [S0021-8979(98)06618-3]

## I. INTRODUCTION

Recently, the most important problem in recording media technology is how to increase the recording density.[1–3] Due to their high coercivity, CoCrM (M=Ni, Ta, Pt) alloy thin films are the most widely used materials in longitudinal recording while CoCr films with columnar grains are used for perpendicular recording. For these crystalline films, the most significant problem is the noise that results at domain transition region from magnetic exchange coupling between the grains.[4] Likewise, intergranular voids, stacking faults, crystallographic orientation, etc., all decrease magnetic performance.[5] Practically, if we want to increase the areal recording density of the crystal film, the grain size of the film must be reduced.[3] However, when the grain size is smaller than the single-domain size, the grains will become superparamagnetic particles and the coercivity of the film will decrease rapidly due to thermal fluctuations. So, the recorded bit size is limited and correlated to the single-domain grain size.

Although the fabrication of single-domain nanocrystal particles might be achieved by various methods,[6–9] a Gaussian distribution of particle size may occur during the fabrication of thin films. It is difficult to obtain uniform single-domain particles. This distribution of grain size results in some of the particles being multidomain and some of the others will be superparamagnetic. Moreover, the location of and distance between the particles is uncontrollable.

In this article we suggest that the high coercivity amorphous thin films may be one of the most promising candidates for ultrahigh density magnetic recording (areal recording density $>50$ Gbits/in$^2$) both in longitudinal recording and perpendicular recording.

Amorphous Co–Tb alloy thin films have excellent qualities of composition modulation and special perpendicular anisotropy. They have been widely studied by many investigators.[10–12] Amorphous thin films of TbFeCo have long been used as recording media in Magnetooptical (MO) data storage.[13–15] If the wavelength of the recording laser beam is small enough, the record spots in this amorphous film might be reduced to a uniformly isolated single-domain size with desired shape and smallest distance between neighboring recorded bits without grain boundary or crystallographic orientation problems.

In this work, dc magnetron sputtering was used to prepare high coercivity amorphous Co–Tb thin films that might be used in ultrahigh density magnetic recording. The effects of composition, sputtering power, argon pressure, and annealing temperature on the perpendicular and in-plane magnetic properties of the Co–Tb thin films were investigated. A nearly magnetically isotropic amorphous Co–Tb film with in-plane coercivity of about 2080 Oe was obtained after annealing. The maximum perpendicular coercivity was as high as 6000 Oe for the as-deposited amorphous films.

## II. EXPERIMENT

Amorphous $Co_{100-x}Tb_x$ ($x=7-60$) alloy thin films were produced by dc magnetron sputtering. A mosaic target consisting of high purity cobalt disk (99.99%) overlaid with high purity terbium pieces (99.9%) was used. By varying the number of Tb pieces arrangement provides for a wide range of effective target compositions and therefore film compositions. The films were deposited onto glass substrates at room temperature.

Applied dc power source was working at controlled various powers and various deposition rates. The base pressure was $5\times10^{-7}$ Torr and after the high purity Ar gas was introduced, various sputtering pressures were investigated. 1000-Å-thick Co–Tb films were used in this study. To protect the film from oxidation, the magnetic layer was sandwiched between a $SiN_x$ layer and the glass substrate. The 500-Å-thick protective $SiN_x$ layer was sputtered from a Si wafer target by dc magnetron reactive sputtering. The sputtering pressure was 5 mTorr with a mixture of Ar and $N_2$ gases.

The magnetic properties of the films were measured with a vibrating sample magnetometer (VSM) at room tempera-

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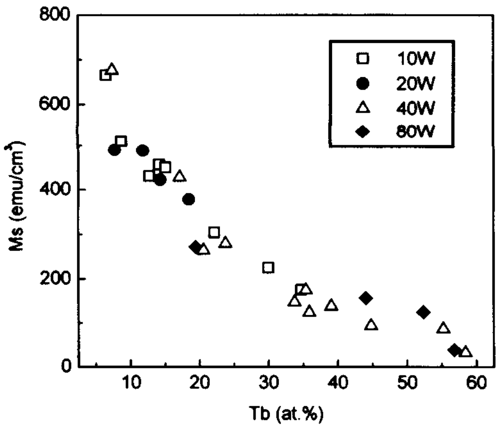


FIG. 1. Variation of saturation magnetization Ms at room temperature of the as-deposited amorphous Co–Tb film as a function of Tb content. The sputtering power was varied from 10 to 80 W. The Ar pressure was 3 mTorr.

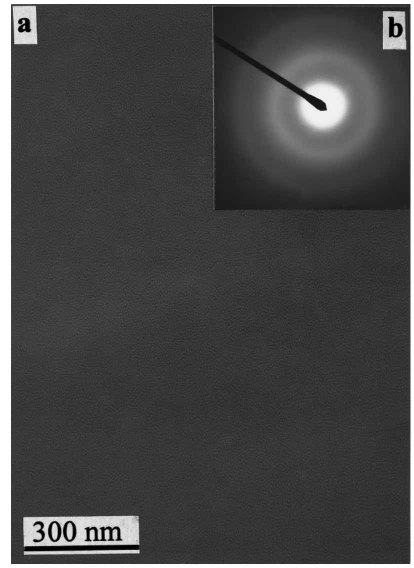


FIG. 2. TEM photo and diffraction pattern of the as-deposited $Co_{64}Tb_{36}$ film. (a) the TEM shows a lemon peel-like microstructure and (b) the diffraction pattern indicates an amorphous structure.

ture using a maximum applied field of 12 kOe. The microstructure of the films was characterized by transmission electron microscopy (TEM). The composition and homogeneity of the films were determined by energy disperse x-ray diffractometer (EDX). The thickness of the films were measured by an $\alpha$ step.

## III. RESULTS AND DISCUSSION

Here, we use different Co–Tb compositions, sputtering powers, argon pressures, and annealing temperatures to examine the variation of the parallel and normal to the film plane magnetic properties.

Figure 1 shows the relationship between the saturation magnetization Ms at room temperature of the as-deposited Co–Tb film at various sputtering powers as a function of the Tb content. The Ar pressure was 3 mTorr. It can be seen that the Ms value was almost independent of the sputtering power but decreases with increasing Tb content. At a Tb content of 7 at. %, the Ms value is 676 emu/cm$^3$. When the Tb content of the film is increased to 58 at. %, the Ms decreases to 30 emu/cm$^3$. This is due to that the amorphous Co–Tb alloy is sperimagnetic/ferrimagnetic,[16] the magnetic moments of Co atoms are antiparallel to that of Tb atoms. The resulting magnetization is the summation of Tb subnetwork magnetization and Co subnetwork magnetization. Due to the larger magnetic moment of Co than that of Tb at room temperature, the resulting Ms value increases with increasing Co content. From Fig. 1, we can see that the compensation composition of the Co–Tb film at room temperature is about $Co_{60}Tb_{40}$. These as-deposited Co–Tb films all have an amorphous structure, as shown in Fig. 2. Figure 2(a) is the TEM photograph of the as-deposited $Co_{64}Tb_{36}$ film. When examined in detail, a lemon peel-like microstructure is observed. The diameter of the lemon peel-like bumps is very small (about 60 Å). The large area diffraction pattern of this lemon peel-like structure yields a broad halo and so the lattice spacing cannot be estimated, as shown in Fig. 2(b). It is obvious that this film has an amorphous structure.

In Fig. 3, the Tb concentration in the Co–Tb films and the deposition rate of Co–Tb films are plotted versus the Ar sputtering pressure ranging from 1 to 20 mTorr. We can see that the value of Tb content increased monotonically with the Ar pressure. The Tb content was 36 at. % in the film when the Ar pressure was 1 mTorr. It increased to 44 at. % as the Ar pressure increased to 20 mTorr. The deposition rate was about 4 Å/s when the Ar pressure was less than 5 mTorr. It decreased rapidly when the Ar pressure was higher than 5 mTorr. The deposition rate decreased to 2.3 Å/s at 10 mTorr then remained constant when the Ar pressure was higher than 10 mTorr. So, in the following we selected Ar pressures of 3 and 5 mTorr to examine the effect of the other sputtering parameters on the magnetic properties of the films.

Figure 4 shows the relationship between coercivity Hc and Tb content of the as-deposited films. The sputtering power was 40 W and Ar pressure was 3 mTorr. The in-plane coercivity $Hc_{\parallel}$ seems to be independent on the Tb content and is less than 400 Oe over the whole composition range. The perpendicular coercivity $Hc_{\perp}$ was also lower than 200

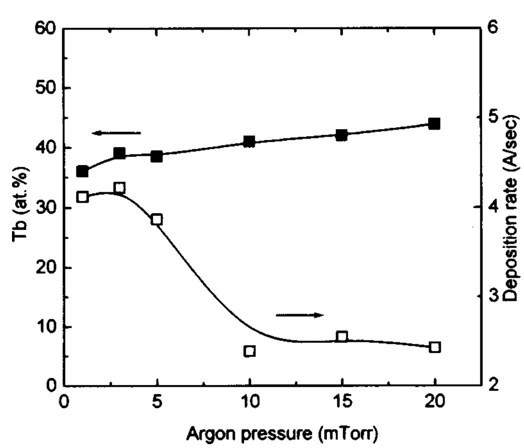


FIG. 3. The correlation between the Tb content of the film, deposition rate, and Ar pressure. The sputtering power was fixed at 40 W.

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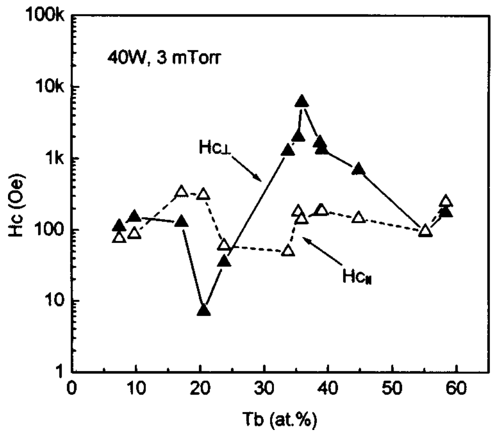


FIG. 4. The variation of coercivity with Tb content for the as-deposited Co–Tb films. The dash line is for the in-plane coercivity and solid line is for the perpendicular coercivity. The sputtering power was 40 W and the Ar pressure was 3 mTorr.

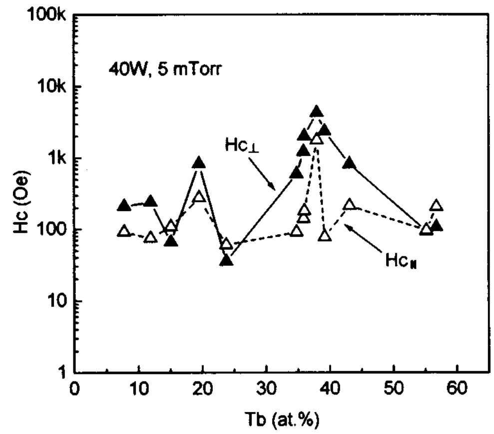


FIG. 5. The variation of coercivity with Tb content for the as-deposited Co–Tb films. Dash line is for the in-plane coercivity and solid line is for the perpendicular coercivity. The sputtering power was 40 W and the Ar pressure was 5 mTorr.

Oe when the Tb content was below 25 at. % and above 50 at. %. The maximum $Hc_\perp$ occurred in the composition range between $Co_{65}Tb_{35}$ and $Co_{63}Tb_{37}$. The $Hc_\perp$ was higher than 6000 Oe, when the Tb content was 36 at. % (i.e., $Co_{64}Tb_{36}$). Its Ms value is about 130 emu/cm$^3$, as shown in Fig. 1.

Figure 5 shows the relationship between the coercivity and the Tb content for the as-deposited Co–Tb films as the Ar pressure was increased to 5 mTorr. For these films, the in-plane coercivity $Hc_\parallel$ and perpendicular coercivity $Hc_\perp$ almost have a similar variation over the entire composition range. When the Tb content is between 36 and 39 at. %, $Hc_\perp$ is higher than 1000 Oe. The maximum $Hc_\perp$ is about 4230 Oe and the maximum $Hc_\parallel$ is about 1770 Oe. The maximums both occur at 38 at. % Tb (i.e., $Co_{62}Tb_{38}$). Comparing Fig. 4 with Fig. 5, we can see that $Hc_\perp$ and $Hc_\parallel$ vary with the change of Ar pressure. As the Ar pressure is increased from 3 to 5 mTorr the maximum $Hc_\perp$ of the film is decreased from 6000 to 4230 Oe. But $Hc_\parallel$ increases from 200 to 1770 Oe when the Tb content is 38 at. %. The magnetic anisotropy of the film changes from perpendicular to isotropic; this may be due to the variation of the microstructure caused by the increase in Ar pressure.[17]

In Fig. 6, the parallel and normal to the film plane squareness Mr/Ms of the as-deposited $Co_{62}Tb_{38}$ films are plotted versus the Ar sputtering pressure in the range from 1 to 20 mTorr. As the Ar pressure increases from 3 to 5 mTorr we can see that the in-plane squareness increases from 0.02 to 0.61, but the perpendicular squareness decreases from 0.83 to 0.64. When the Ar pressure is higher than 5 mTorr, the in-plane and perpendicular squareness both decrease with increasing Ar pressure. The maximum perpendicular squareness and minimum in-plane squareness both occur at 3 mTorr of Ar pressure. This indicates that when the Ar pressure is 3 mTorr, the magnetic easy direction is normal to the film plane. This is the reason why we select Ar pressure as 3 mTorr to prepare perpendicular magnetic anisotropy films and Ar pressure is 5 mTorr for preparing magnetically isotropic Co–Tb films.

Figure 7 shows the coercivity and squareness of the $Co_{62}Tb_{38}$ film as a function of annealing temperature Ta between 100 and 250 °C. The annealing time was 60 min. These films were deposited at a dc sputtering power of 40 W and Ar pressure of 5 mTorr. When Ta is higher than 250 °C, very small crystalline particles appear and the magnetic properties of the film decrease rapidly. This is consistent with the reported crystallization temperature of 255–300 °C[16] for the evaporated amorphous Co–Tb films. Figure 7(a) shows the in-plane coercivity $Hc_\parallel$ and squareness $S_\parallel$. When Ta is increased from 100 to 180 °C, the $Hc_\parallel$ value remains nearly constant at about 1600 Oe and the $S_\parallel$ stays at about 0.54. But, as Ta increased to greater than 180 °C, $Hc_\parallel$ and $S_\parallel$ both increase with increasing Ta. $Hc_\parallel$ increases from 1600 to 2080 Oe and $S_\parallel$ increases from 0.54 to 0.8 as Ta increases from 180 to 250 °C. Figure 7(b) shows the perpendicular coercivity $Hc_\perp$ and squareness $S_\perp$. When Ta is increased from 100 to 200 °C the $Hc_\perp$ value almost remains constant at about 4100 Oe and the $S_\perp$ value stays at about 0.77. As Ta is higher than 200 °C, $Hc_\perp$ decreases rapidly from 4100 Oe at Ta=200 °C to 2400 Oe at Ta=220 °C then

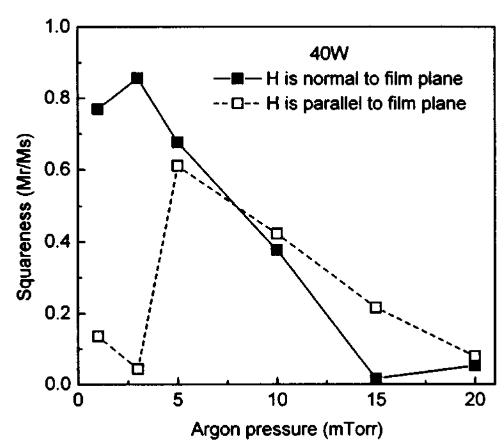


FIG. 6. Squareness Mr/Ms of as-deposited $Co_{62}Tb_{38}$ film vs Ar sputtering pressure. The sputtering power is 40 W. The solid line and the dash line represent the applied field H is normal and parallel to the film plane, respectively.

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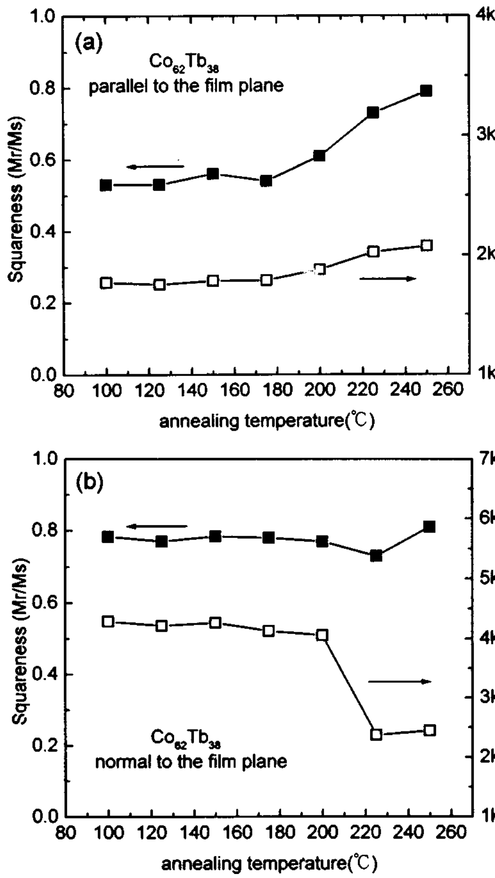


FIG. 7. Squareness and coercivity dependence on annealing temperature for $Co_{62}Tb_{38}$ films. The annealing time was 60 min, (a) is the in-plane squareness and coercivity, and (b) is the perpendicular squareness and coercivity.

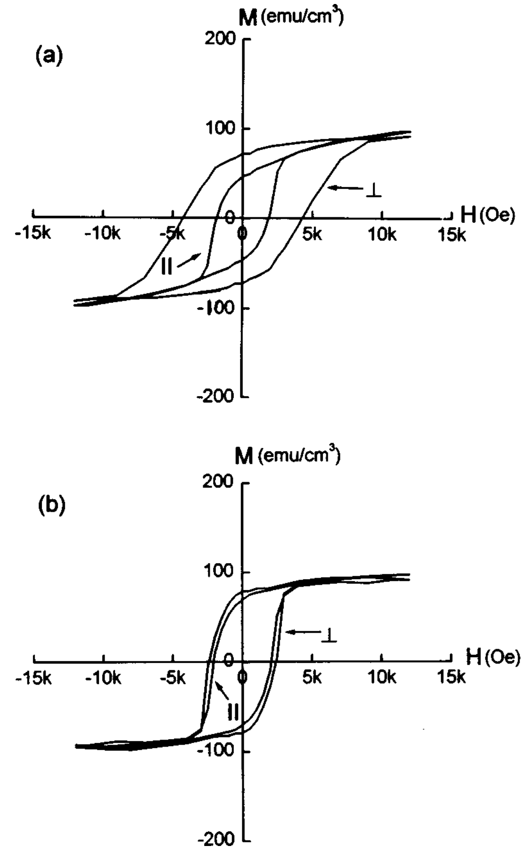


FIG. 9. M–H loops measured along the directions of normal (⊥) and parallel (∥) to the film plane of the $Co_{62}Tb_{38}$ film. (a) As deposited, (b) After annealing in vacuum at 250 °C for 60 min.

keeps constant when Ta is higher than 220 °C. But, $S_{\perp}$ increases from 0.75 to 0.82 as Ta increases from 220 to 250 °C. After annealing, we can see that the magnetic anisotropy of the films is changed due to the stress release. The $Co_{62}Tb_{38}$ films which were annealed between 220 and 250 °C almost have isotropic magnetic properties. These annealed $Co_{62}Tb_{38}$ films also have an amorphous structure, as shown in Fig. 8. Figure 8(a) is the TEM photograph of the $Co_{62}Tb_{38}$ film after being annealed at 250 °C. It also has a lemon peel-like microstructure. The diameter of these lemon peel-like bumps is about 37 Å which is smaller than that of Fig. 2(a). The broad halo diffraction pattern, as shown in Fig. 8(b), indicates that this film is also an amorphous structure.

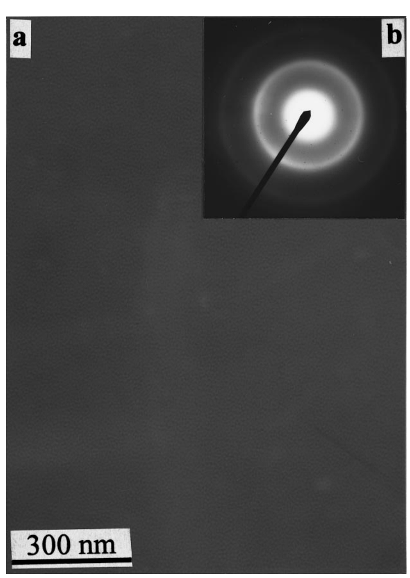


FIG. 8. TEM photo and diffraction pattern of the $Co_{62}Tb_{38}$ film which was annealed at 250 °C for 60 min, (a) is the TEM microstructure and (b) is its diffraction pattern.

Figure 9(a) shows the M–H curves of the as-deposited $Co_{62}Tb_{38}$ film. This film was deposited at dc power of 40 W and Ar pressure of 5 mTorr. For this film, the in-plane coercivity is 1770 Oe and the perpendicular coercivity is 4230 Oe. The Ms value of this film is about 100 emu/cm$^3$. Figure 9(b) shows the M–H curves of this film after it was annealed at 250 °C for 60 min. The M–H curves show that the film exhibits nearly isotropic magnetic properties. The in-plane coercivity is 2080 Oe and the perpendicular coercivity is 2460 Oe.

## IV. CONCLUSIONS

The magnetic properties of sputtered amorphous $Co_{100-x}Tb_x$ alloy thin films have been studied over a wide composition range, sputtering power, and argon pressure. An amorphous structure is still obtained, but with isotropic magnetic properties for the $Co_{62}Tb_{38}$ films annealed at 250 °C for 60 min in vacuum. The in-plane coercivity was about 2080

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Oe and the Ms value was about 100 emu/cm$^3$. For the as-deposited amorphous $Co_{100-x}Tb_x$ films with $x = 35-39$ at. % the perpendicular coercivities are between 2000 and 6000 Oe. The Ms value is about 130 emu/cm$^3$.

## ACKNOWLEDGMENT

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