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Composition and annealing dependence of magnetic properties in amorphous Tb–Co based alloys

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Abstract

The aim of the work is to study the magnetic properties of $Tb_{1-x}Co_x$ amorphous alloys with respect to the composition and annealing temperature. The samples have been prepared by the dc triode co-sputtering. The most important results are: (1) Curie temperature is higher than the room temperature for the magnetic samples with Co content $x>0.6$, before annealing rises T_c with Co content. Crystallization commences at approximately 400°C. (2) A significant high field susceptibility, even up to 8 Tesla, confirms the sperimagnetic structure. (3) A biaxial anisotropy occurs before annealing due to stress, but a uniaxial anisotropy is induced by the field annealing; the anisotropy value is dependent upon composition. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Magnetic films; Amorphous materials; Anisotropy; Magnetization; Amorphization

1. Introduction

The magnetic structure of the rare earth–transition metal amorphous alloys have been intensively studied [1–5]. Among all researched properties, the magnetization and the shape of hysteresis loops are dependent upon the alloy composition, heat and mechanical treatment. Amorphous alloy films are usually prepared by sputtering different procedures.

The random anisotropy model of Harris, Plichke and Zukermann [6] describes the competition between exchange interactions and random anisotropy in amorphous alloys. The model accounts for the sperimagnetism, i.e. the non-colinear arrangement of rare-earth magnetic moments in $3d$–$4f$ alloys due to the large random anisotropy of rare-earth atoms (R). Consequently, the magnetization should be free from anisotropy at large scale and feature high anisotropy at short scale. These amorphous alloys are characterized by large magnetic domains, so-called Imry and Ma domains, which also occurs in the case of strong exchange interaction J and weakly random anisotropy D [7].

In the case of magnetism, the amorphous state changes the magnetic properties due to the narrow relation between crystallographic and magnetic structure. However, in the first approximation [8,9], the mean magnetic moments do not depend upon the surrounding atomic arrangement, but depends upon the chemical composition in either crystallized or amorphous state. As an example, critical content for magnetic disappearance in amorphous is greater than in crystallized.

The aim of the present work is to describe the magnetic properties in the $Tb_{1-x}Co_x$ amorphous alloys with various compositions for an as-cast and after magnetic field annealing. Changes of the composition and of annealing temperature will be considered in the influence upon the magnetization process, the magnetic anisotropy and sperimagnetism. $Tb_{1-x}Co_x$ amorphous thin films are chosen as the system because of its relatively high Curie temperature and its good resistance against oxidation.

2. Experimental

2.1. Preparation of amorphous films

Rare earth –transition metal amorphous films $Tb_{1-x}Co_x$ ($x=0.6$–0.8) samples were prepared by dc triode co-sputtering from various targets onto silica substrates with thicknesses of about 1 μm. The thickness range is from 2000 to 5000 Å. The samples were characterized in crystallographic structure by X-ray diffraction and by Rutherford Back Scattering (RBS) in compositional and

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depth analysis. Heat treatments up to 400°C were carried out on some alloys and their Curie temperature (T_c) was determined.

2.2. Structural analysis by X-ray diffraction

All the samples were characterized by means of 2Θ X-ray diffraction using Kα of copper with λCu=0.15405 nm. These as-sputtered films present the common shape of the X-ray patterns for amorphous alloys. This pattern consist only of broad maxima close to 48° without sharp peaks as described in early work [1,2].

2.3. Compositional analysis by means of RBS

To estimate the thicknesses and chemical composition, the Rutherford Back Scattering was used. The count rate of energy value corresponds to element content at fixed depth of analysis. Stochiometry is deduced from I_{Tb}/I_{Co} widths of peak and gives the thicknesses which were found between 2000 and 5000 Å.

2.4. Magnetization process and hysteresis loops

The basic magnetic properties of the samples were measured by a VSM in fields up to 8 Telsa. We noted firstly that all samples with cobalt content x less than 0.6 have their Curie temperatures below room temperature (300 K). T_c increases with Co content x as shown in Fig. 1. For $Tb_{0.31}Co_{0.69}$, T_c is about 520 K, as shown in inset of Fig. 1. The magnetization and magnetic anisotropy were measured at 20 and 300 K. Hysteresis loops allowed to determine the principal characteristics of magnetization.

3. Results and discussion

3.1. As-cast $Tb_{1-x}Co_x$ films

In the inset of Fig. 2 can be shown the magnetization in high magnetic fields. We can observe the significant high field susceptibility, even up to 8 Tesla. For all samples, complete saturation being unattained. By interpreting Fig. 2, we can observe the dependence of high field susceptibility with composition. At 300 K, for non-magnetic

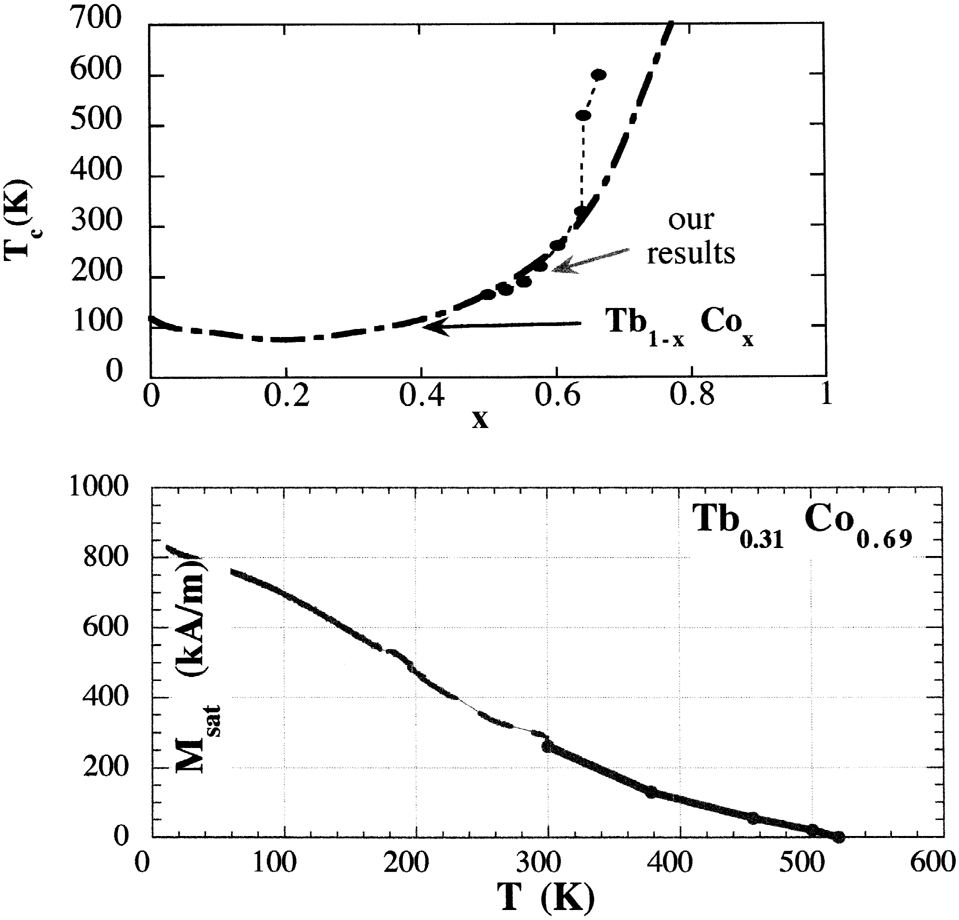

Fig. 1. Compositional dependence of Curie temperature T_c in as-cast $Tb_{1-x}Co_x$ films. Inset: spontaneous magnetization versus temperature in as-cast $Tb_{0.31}Co_{0.69}$ film.

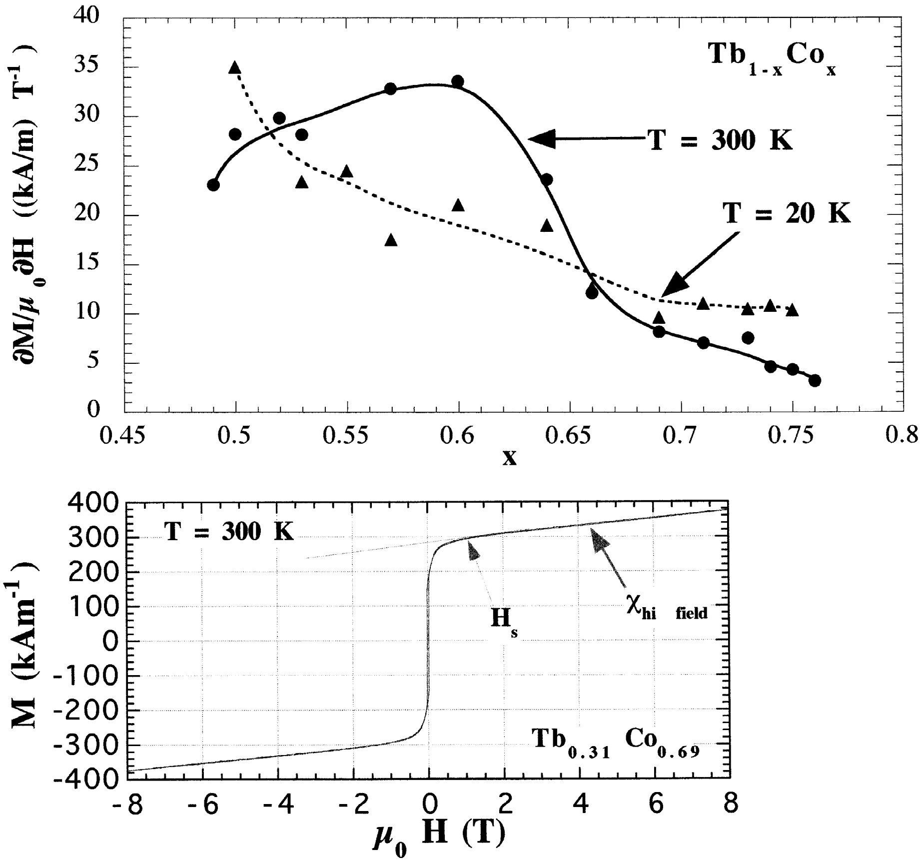

Fig. 2. Compositional dependence of high field susceptibility χ^{M}_{HF} at 20 and 30 K in as-cast $Tb_{1-x}Co_x$ films. Inset: magnetization at high fields versus applied field in $Tb_{0.31}Co_{0.69}$ film.

samples ($x<0.6$), χ^{M}_{HF} increases with x until T is close to T_c ($x=0.6$) and χ^{M}_{HF} is purely paramagnetic. When x is more than 0.6, χ^{M}_{HF} decrease due to increasing of $J_{Co-Tb} \gg H_{applied}$. Also, in the low temperatures, random anisotropy dominates because $J_{Co-Tb} \gg D$.

With regard to Fig. 3, which shows compositional dependence of spontaneous magnetization Ms (extrapolated at 0 Tesla) at 300 K, $x=0.8$ corresponds to the compensation point between magnetic moments of Tb and Co. This value of concentration rate corresponds to the disappearance of magnetism. Fig. 3 illustrates the mean value of moment along the magnetic field H, both below and above compensation point.

The samples which are magnetic at 300 K show hysteresis loops to be characteristic of an anisotropy which is dependent on composition, as shown in Fig. 4a and b. The loops were measured along easy and hard magnetic axes in plane of film. Anisotropy depends upon composition: for low Cobalt content (Fig. 4a), the magnetization lies in the plane of the sample with a very small

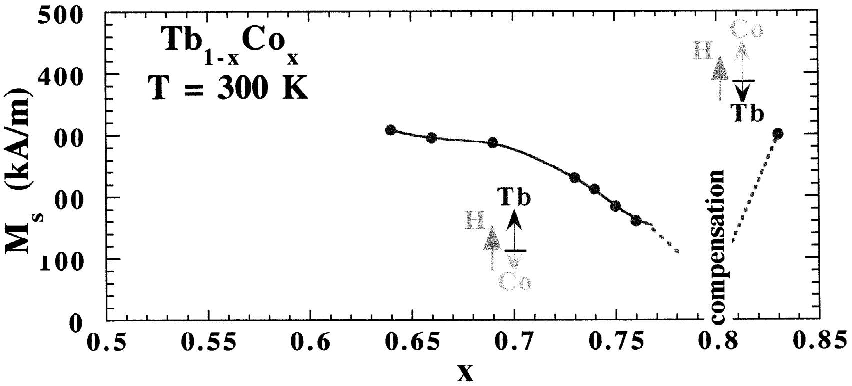

Fig. 3. Compositional dependence of spontaneous magnetization at 300 K in as-cast $Tb_{1-x}Co_x$ film.

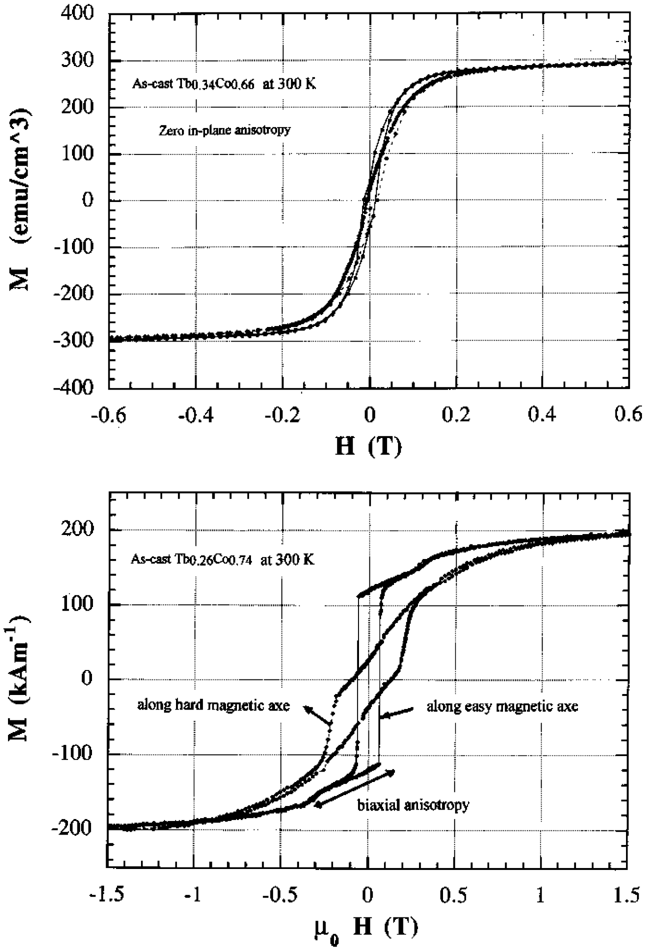

Fig. 4. Hysteresis loops at 300 K in: (a) in as-cast $Tb_{0.34}Co_{0.66}$ film; and (b) in as-cast $Tb_{0.26}Co_{0.74}$ film.

anisotropy (small M_R/M_S). On the contrary, for higher Co content (Fig. 4b), competition occurs between a well defined in-plane anisotropy and a perpendicular anisotropy. This is a biaxial anisotropy regime. Anisotropy and coercive field of these samples rapidly increases with x at 300 K (see Fig. 6a and b).

Regarding Fig. 4b, the susceptibility in strong fields shows alignment of moments in the applied field direction. In weak fields, the competition between local crystalline fields and exchange interactions leads to the occurrence of Imry and Ma domains of size 10^{-7} m for $TbCo_3$ [3]. An irreversible reorganisation of these domains under applied fields is revealed in coercivity measurements.

3.2. Annealed $Tb_{1-x}Co_x$ films

Some samples with x ranging from 0.6 to 0.75 were annealed for 1 h at different temperatures from Ta=75 to 400°C under applied field about 2.5 Tesla applied perpendicular to the length in the film plane.

X rays indicate that the crystallization starts above 350°C. Peaks of Tb, Co_2 and Co appear on contrary of the broad maxima in as-cast samples.

Regarding Fig. 5, we can observe the change of hysteresis loops at 300 K before and after annealing in $Tb_{0.31}Co_{0.69}$: Firstly, the shape of loop in the annealed sample shows a zero plane anisotropy on contrary of as-cast sample which shows a biaxial anisotropy. Secondly, increasing magnetization with reducing a high field susceptibility and the coercive field. As shown in Fig. 6a and b, coercive field Hc and anisotropy K (M_R/M_S) are reported. For lower x-values, low annealing temperature Ta is efficient to change magnetic properties Hc and K (anisotropy). For higher x-values, higher Ta is required. Stress are relaxed by annealing, which leads to the rapid decrease of Hc and the induced uniaxial anisotropy by the field annealing.

4. Conclusion

We have studied the series of amorphous $Tb_{1-x}Co_x$ with various compositions. As-cast samples showed a high T_c which rises with Co content. The sperimagnetism of this alloys are confirmed which are hard magnetic and characterized by a biaxial anisotropy in the film plane. Field

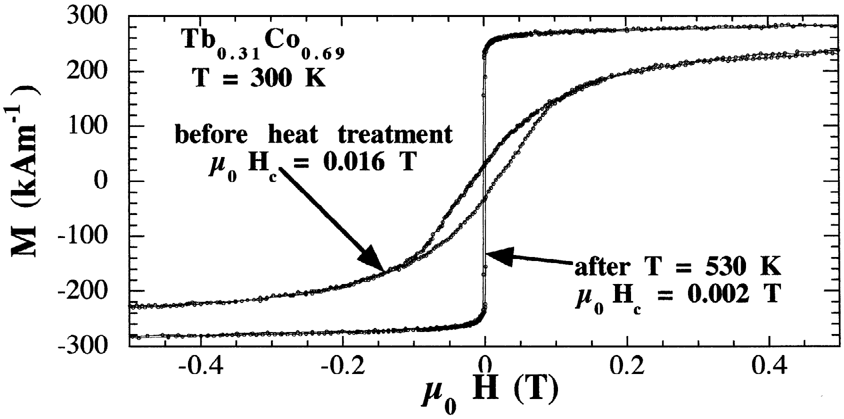

Fig. 5. Hysteresis loops at 300 K in an as-cast and in an annealed $Tb_{0.31}Co_{0.69}$ film.

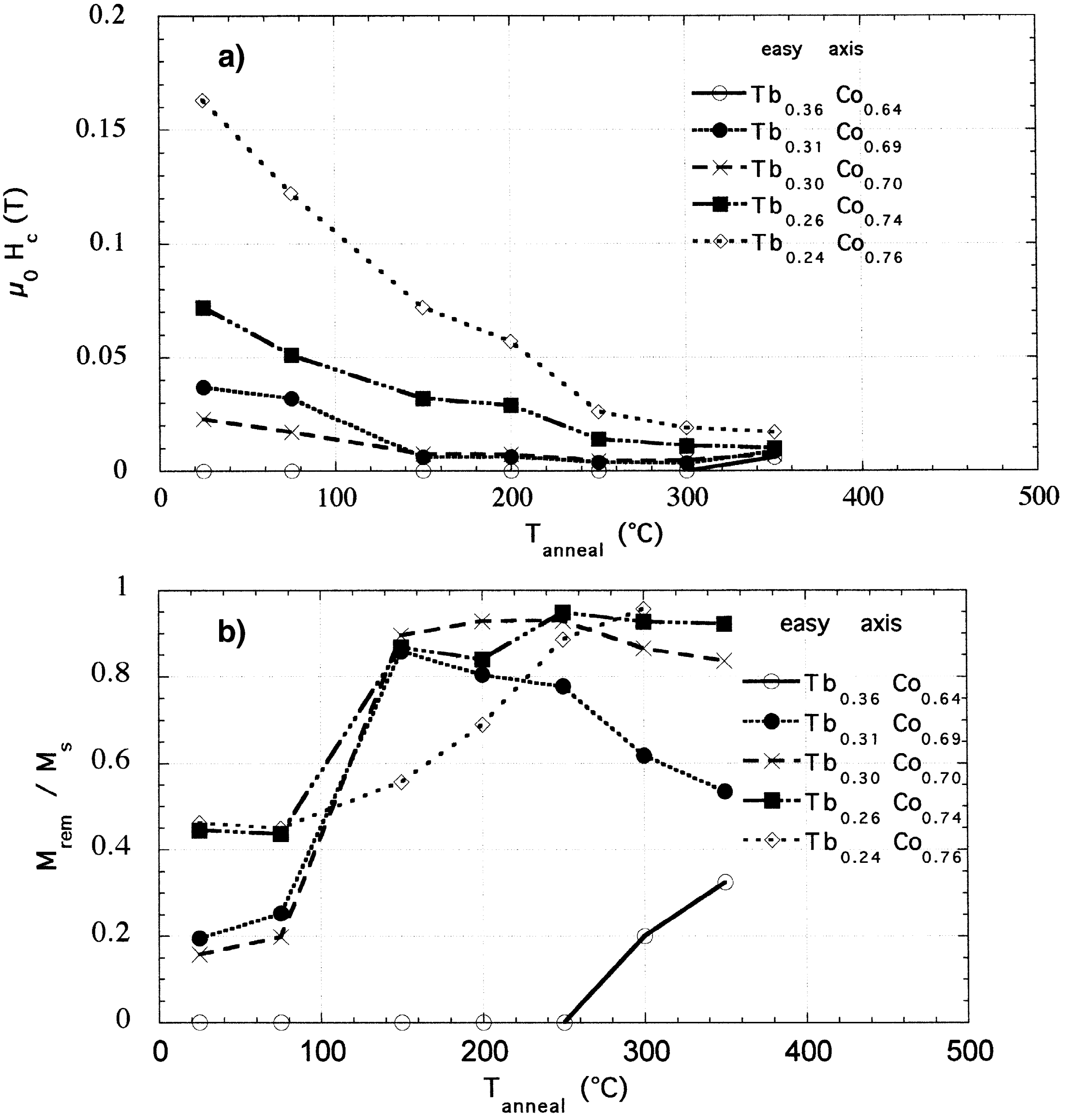

Fig. 6. Annealing dependence for some samples along easy axis ($0.60<x<075$) of: (a) coercive field; and (b) anisotropy.

annealing make the samples magnetically softer. With a sufficient field annealing has desired effects: firstly, by reducing the internal stress which are induced during the deposition process; secondly, by inducing a well-defined in plane anisotropy.

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