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Enhanced coercive force of Nd–Fe–B thin films by the introduction of a Cr underlayer

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Abstract

We investigated the influence of a Cr underlayer on the structure and magnetic properties of Nd–Fe–B thin films which were produced by a conventional rf sputtering method. The thickness of the underlayer and the substrate temperature were varied from 0 to 5000 Å and from room temperature to 670°C, respectively. It was found that the thickness of the underlayer material and the substrate temperature strongly influenced the crystal orientation and magnetic properties. The main crystalline phase was $Nd_2Fe_{14}B$, whose tetragonal *c*-axis was aligned perpendicularly to the film plane. A high coercive force (H_c=3.7 kOe) was achieved using the following conditions: Cr underlayer with a thickness of 500 Å and a substrate temperature of 650°C. From TEM observation, this enhanced coercive force is thought to be attributable to a modification of the columnar structure.

Keywords: Columnar structure; Hard magnetic material; High coercive force; Nd–Fe–B thin film; Sputtering

1. Introduction

Thin films of hard magnetic Nd–Fe–B alloys are candidate materials for micro-mechanical devices, such as micro-motors and actuators, and data storage media. In 1986, Cadieu et al. reported that oriented $Nd_2Fe_{14}B$ films could be produced by sputtering [1]. Since then, many efforts have been made on these hard magnetic thin films [2,3]. However, most work has been performed on the influence of the substrate temperature, the adjustment of the composition and the preparation refinement using an UHV sputtering method in order to avoid oxidation of the thin film. On the other hand, the introduction of a Cr underlayer to Sm–Co thin films has been reported [4,5]. From the results for the Sm–Co system, the introduction of an underlayer material is very effective in modifying the morphology of the columnar structure and in enhancing the coercive force. Recently, several studies dealing with the effects of underlayer materials have been reported [6–8]. However, the relationship between the magnetic properties and the microstructure has not yet been clearly elucidated.

In this study, we investigated the influence of a Cr underlayer, its thickness and substrate temperature on the structure and, consequently, the magnetic properties of Nd–Fe–B thin films.

2. Experimental procedure

Nd–Fe–B thin films were prepared using a conventional rf sputtering method. The thickness of the Nd–Fe–B layer was fixed at 3000 Å while the thickness of the Cr underlayer was varied from 0 (without underlayer) to 5000 Å. The top of the Nd–Fe–B layer was coated with a protective layer with a composition identical to the underlayer. $Nd_{13}Fe_{70}B_{17}$ (at.%) and 99.9 at.% Cr disks were used as the target. The substrate used in the present study was glass (Corning, #7059). The substrate temperature was varied in the range from room temperature to 670°C. The base pressure was $<7.0\times10^{-7}$ Torr and the Ar gas pressure during sputtering was 7.4 mTorr. The crystalline structure was measured by X-ray diffractometry with Cu Kα radiation and, in some cases, by high-resolution transmission electron microscopy. A focused ion beam apparatus (SMI9200, Seiko Instruments Inc.) was used for preparing the cross-sectional specimens. Coercive force and saturation magnetization were measured using a VSM

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at fields up to 15 kOe and also by a SQUID magnetometer at fields up to 55 kOe. The film composition was determined by ICP spectrometry.

3. Results and discussion

Fig. 1 shows the magnetic hysteresis loops at various substrate temperatures from room temperature to 670°C. The loops were measured in directions both parallel (//) and perpendicular (⊥) to the film plane. The film deposited at room temperature exhibits anisotropic magnetization. The easy direction is parallel to the film plane and the film is magnetically soft (H_c<30 Oe). However, with increasing substrate temperature, the easy direction of magnetization changes from parallel to perpendicular and a large coercive force is obtained at 650°C. At temperatures higher than 670°C, the coercive force measured in the parallel direction is increased. This similar behavior of the relationship between the substrate temperature and the magnetic properties has also been reported previously in the literature.

Nd–Fe–B thin films deposited on the room temperature substrate have large-angle X-ray diffraction patterns, characteristic of an amorphous state. On the other hand, peaks from the tetragonal *c*-axis of $Nd_2Fe_{14}B$ are observed for the films deposited at substrate temperatures above 580°C.

Fig. 2 shows the X-ray diffraction patterns for [Cr(*t* Å)/Nd–Fe–B(3000 Å)/Cr(500 Å)] thin films with various Cr underlayer thicknesses (*t*=0, 100, 500 and 5000 Å). All samples were deposited at the substrate temperature of 650°C. From this figure, only the peaks from the tetragonal *c*-axis of $Nd_2Fe_{14}B$ are seen in the thickness range 0–500 Å. In the case of the Cr underlayer thickness of 5000 Å the peaks from different planes begin to appear. It is thought that the greater the thickness of the underlayer, the more the crystallographic orientation collapses. It is therefore considered that the underlayer thickness strongly influences the crystal orientation of Nd–Fe–B thin films.

Fig. 3 shows a series of hysteresis loops for [Cr/Nd–Fe–B/Cr] thin films with various thicknesses of Cr underlayer. The Nd–Fe–B thin films deposited directly onto the glass substrate at 650°C exhibit anisotropic magnetization whose easy direction is perpendicular to the film plane. Among these samples, the film with *t*=500 Å shows a large coercive force in the direction perpendicular to the film plane. However, the film with *t*=5000 Å shows a fairly large coercive force in the direction parallel to the film plane.

Fig. 4 shows the effect of the thickness of the Cr underlayer on the magnetization and coercive force for [Cr/Nd–Fe–B/Cr] thin films. Solid circles and triangles denote the data obtained from the perpendicular magnetization and the in-plane magnetization, respectively. All samples were deposited at a substrate temperature of 650°C. The saturation magnetization of these films shows a

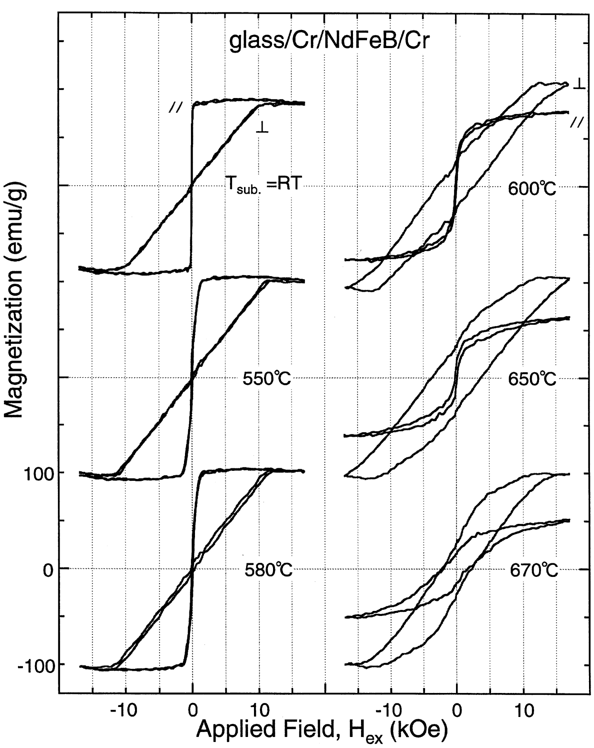

Fig. 1. Magnetic hysteresis loops at various substrate temperatures. The substrate temperature is varied from room temperature to 670°C.

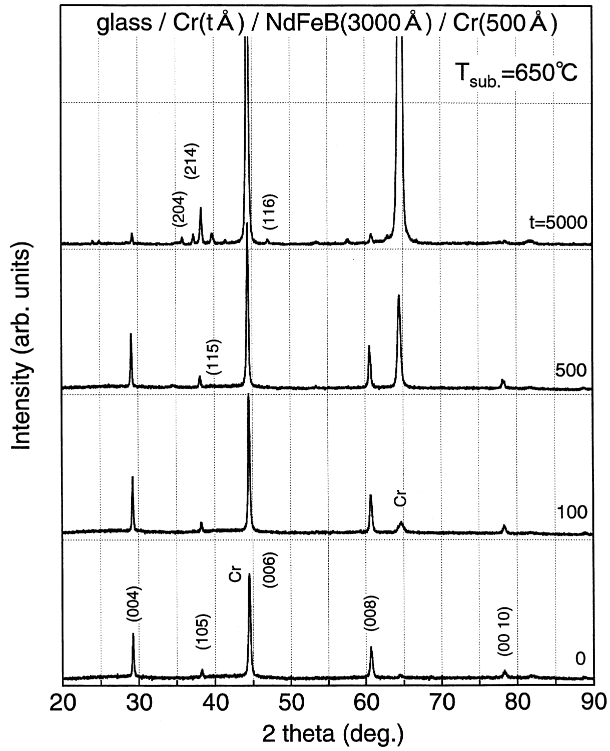

Fig. 2. X-ray diffraction patterns for [Cr(*t* Å)/Nd–Fe–B(3000 Å)/Cr(500 Å)] thin films with various Cr underlayer thicknesses (*t*=0, i.e. without underlayer, 100, 500 and 5000 Å).

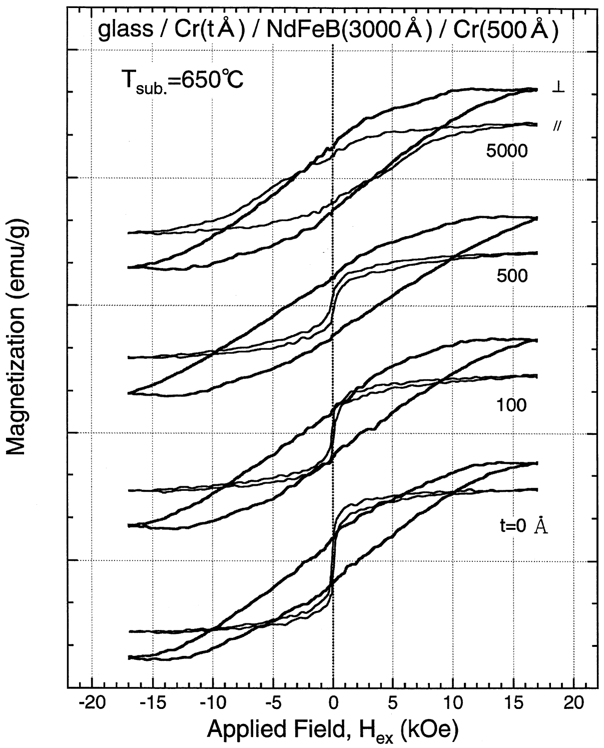

Fig. 3. Room temperature hysteresis loops for [Cr/Nd–Fe–B/Cr] thin films with various Cr underlayer thicknesses (*t*).

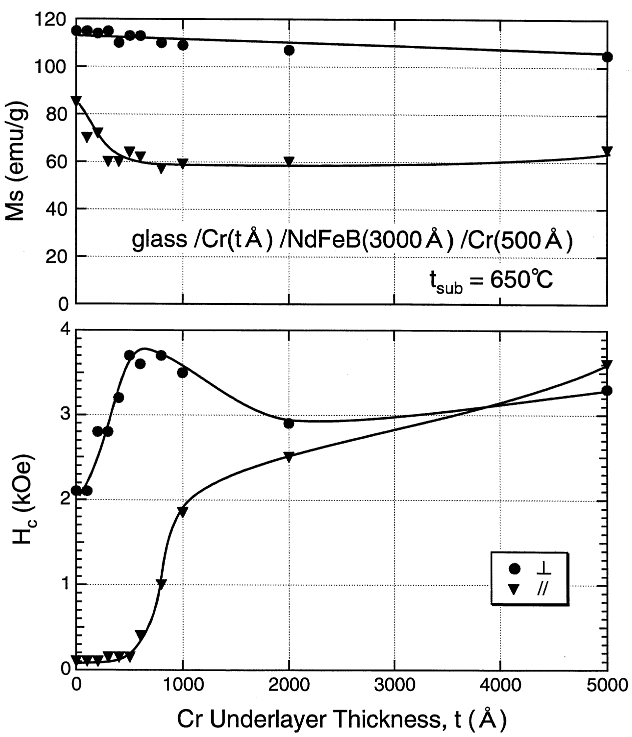

Fig. 4. Effects of the Cr underlayer thickness (*t*) on the magnetization and coercive force of [Cr/Nd–Fe–B/Cr] thin films.

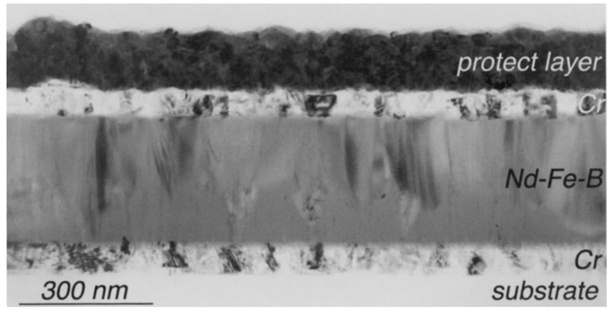

Fig. 5. Cross-sectional TEM image of [Cr/Nd–Fe–B/Cr].

nearly constant value of about 110 emu g^{-1}, although the underlayer thickness is varied. However, the coercive force measured in the perpendicular direction increases with increasing underlayer thickness, reaching a maximum value of 3.7 kOe at a thickness of 500 Å. The coercive force measured in the parallel direction shows a small value below a thickness of 500 Å but begins to increase as the underlayer thickness exceeds 500 Å. A very large change in the coercive force is observed at a thickness of about 500 Å, as can be seen from Fig. 4. Therefore, it is of interest to examine the microstructure in this thickness region.

Fig. 5 shows the cross-sectional TEM image of a Nd–Fe–B thin film with a Cr underlayer thickness of 500 Å. This TEM specimen was fabricated by an FIB apparatus and the thickness of the fabricated sample was less than 700 Å. The protective layer shown at the top of the film was coated in order to avoid damage from Ga ion bombardment from the FIB during sample preparation.

It is of interest to see that the Nd–Fe–B thin film consists of two phases: one is crystalline $Nd_2Fe_{14}B$ with a columnar structure and the other is an amorphous phase. The amorphous state still exists at a substrate temperature of 650°C. It is thought that the enhanced coercive force of this Nd–Fe–B thin film is related to the morphology of the columnar structure. In order to achieve a large coercive force, it is thus important to reduce the remaining amorphous phase and, simultaneously, to control the formation of the columnar structure.

4. Conclusions

The microstructure and magnetic properties of Nd–Fe–B thin films have been found to vary sensitively with the thickness of the Cr underlayer as well as substrate temperature. It has also been found that a large coercive force magnitude can be achieved at a substrate temperature of 650°C. The main phase is $Nd_2Fe_{14}B$, whose tetragonal *c*-axis is aligned in the direction perpendicular to the film plane. The coercive force measured in the direction perpendicular to the film plane ($H_{c\perp}$) increases with increasing underlayer thickness and reaches a maximum value of 3.7 kOe at a thickness of about 500 Å. From the results of TEM observation, it is thought that the enhanced coercive force of this Nd–Fe–B thin film can be attributed to modification of the columnar structure.

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