Exchange biasing in as-prepared Co/FeMn bilayers and magnetic properties of ultrathin single layer films

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Received 14 August 2004; accepted in revised form 22 March 2005
Available online 10 May 2005

Abstract

For Co and CoNi single layer films, the coercivity is a linear function of the inverse layer thickness and has an angular dependence of a fourfold-like symmetry although only a uniaxial anisotropy exists. For both as-prepared and post field-cooled Co/FeMn bilayers, the exchange field and the coercivity enhancement are proportional to the inverse Co thickness. For as-prepared ones, the exchange field and the coercivity are smaller than those of post field-cooled ones. At the same time, the exchange field, the coercivity, and the in-plane resonance field have the similar angular dependence as those of post field-cooled ones. The magnetization reversal process must be accompanied by non-coherent rotation model in Co and CoNi single layer films, and as-prepared Co/FeMn bilayers.

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PACS: 75.30.Et; 75.30.Gw; 75.60.Jk; 75.70.Cn
Keywords: Exchange biasing; Ferromagnetic; Antiferromagnetic

1. Introduction

When exchange coupling is established in ferromagnet(FM)/antiferromagnet (AFM) bi-layers, the hysteresis loop of the FM layers will shift away from the zero magnetic field and the coercivity $H_C$ will usually be enhanced in comparison with corresponding free FM layer [1–3]. Basic features have been extensively studied. First, the shift amount, i.e., the exchange bias field $H_E$ is proportional to $1/t_{FM}$, where $t_{FM}$ is the FM layer thickness. The enhanced $H_C$ decreases with increasing $t_{FM}$. Secondly, so-called negative isotropic in-plane (ferromagnetic) resonance field shift $H_{ISO}$ exists.

In order to clearly investigate the nature of the exchange biasing, FM/AFM bilayers are normally cooled down from high temperatures to low temperatures under an external magnetic field, which is large enough to saturate the samples. As well known, however, the exchange biasing in FM/AFM bilayers in spin valve giant magnetoresistance (GMR) and magnetic tunneling junction (MTJ) devices is established by the deposition field instead of normal field cooling (FC) procedure after deposition [4]. $H_E$ and $H_C$ in as-prepared FM/AFM bilayers are generally identified by magnetometer measurements and GMR curve. Few reports have appeared on the characteristics of the exchange biasing in as-prepared samples. For as-prepared permalloy/FeMn bilayers, the $H_E$ and the enhanced $H_C$ are much smaller than those with post-FC procedure [5]. Systematic studies are required about the characteristics of the exchange biasing in as-prepared bilayers since it is important to fabrications of the GMR and MTJ devices.

There is lack of investigations of magnetic properties of ultrathin FM layer films. Systematic studies are required. First, in most of studies about the exchange biasing, magnetic properties of corresponding free FM layers have been neglected although they might have influence on the exchange biasing of FM/AFM bilayers. Secondly, it is also important to design of MTJ devices such as magnetic
random access memory because ultrathin magnetic layers are widely used in MTJ devices and the magnetization reversal process has great influence on the noise level of these devices [6]. Finally, the mechanism of \( H_C \) for ultrathin single layer films is less well understood although the dependence of \( H_C \) on \( t_{FM} \) has been studied before [6–10]. The variation trend of \( H_C \) with \( t_{FM} \) can be divided into two categories. Some experiments showed that \( H_C \) increases with increasing \( t_{FM} \). Other experiments, however, showed that \( H_C \) decreases with increasing \( t_{FM} \). In this paper, we will first report on magnetic properties of Co and CoNi single layer films. Secondly, we will study the exchange biasing of as-prepared and post-FC Co/FeMn bilayers. To establish the exchange biasing by the post-FC procedure, the samples were cooled from 440 K to room temperature under an external magnetic field of 1.0 kOe, which is aligned parallel to the deposition field.

2. Experimental details

All the samples (1 cm × 5 cm) were prepared at ambient temperature by DC and RF magnetron sputtering on Si(100) substrates, including Si(100)/Cu(30 nm)/CoO(30 nm)/CoNi(0–30 nm)/Cu(30 nm) and Si(100)/Ag(15 nm)/Co(0–10 nm)/Ag(30 nm), and Si(100)/Ag(15 nm)/Co(0–10 nm)/FeMn(20 nm)/Ag(30 nm). These samples are denoted as CoNi and Co single layer films, and Co/FeMn bilayers. Metallic layers were made from corresponding metallic or alloying targets by DC magnetron sputtering and CoO layers were made from CoO target by RF magnetron sputtering. The base pressure was 2 × 10^{-5} Pa and the Ar pressure 0.33 Pa during deposition. The growth rates were 0.1–0.4 nm/s. Ag buffer layers with 15 nm thickness were used to promote the growth of AFM FCC FeMn with preferred (111) orientation. All layers had uniform thickness except the wedge Co and CoNi layers with the wedge shape along the long side of the rectangle. In this way, we can alleviate the run-to-run variation commonly encountered in individually fabricated samples. For all samples, a magnetic field of about 130 Oe was applied parallel to the film plane and along the wedge direction to induce an in-plane uniaxial anisotropy in Co single thick films and unidirectional anisotropy in Co/FeMn bilayers. Microstructure of Co single layer films and Co/FeMn bilayers were analyzed by X-ray diffraction. The structural properties are shown in Fig. 1. One can find the diffraction peaks of FCC Ag(111), Ag(200) and Co(111). FeMn(111) diffraction peak might overlap with those of Co(111) and Ag(200). No other diffraction peaks are found for FeMn layers for Co/FeMn bilayers.

Before magnetic measurements, each large specimen was cut into many small pieces along the wedge direction, thus providing many small samples which had been prepared at the same time but having various \( t_{FM} \). To analyze the magnetic properties, in-plane hysteresis loops were measured by vibrating sample magnetometer (VSM). Ferromagnetic resonance (FMR) measurements were carried out at room temperature with a Bruker ER 200D-SRC electronic paramagnetic resonance spectrometer operating at a fixed microwave frequency of 9.78 GHz and a swept external DC field. FMR spectra at various orientations of the DC magnetic field were obtained by changing the azimuthal angle in the film plane.

3. Results and discussion

Fig. 2 shows typical hysteresis loops along the easy axis for Co single layer films, as-prepared and post-FC Co/FeMn bilayers. It is found that for Co single layer films the hysteresis loops are centered with respect to the zero magnetic field. For as-prepared and post-FC Co/FeMn bilayers, the hysteresis loops are shifted from the zero magnetic field and \( H_C \) is apparently enhanced, in comparison with Co single layer films. With the same \( t_{FM} \), the magnitude of the exchange field \( H_E \) and \( H_C \) of as-prepared samples is smaller than those of post-FC samples.

Fig. 3 shows the \( t_{FM} \) dependence of \( H_C \) along the easy axis for Co and CoNi layer thickness. \( H_C \) of single layer films is a linear function of \( 1/t_{FM} \). Similar experiments were observed before that \( H_C \) of Co ultrathin films was found to decrease with increasing Co layer thickness [9,10]. When the \( 1/t_{FM} \) approaches zero, the intercept is the value of \( H_C \) at infinite \( t_{FM} \). As well known, \( H_C \) is strongly related to the preparation condition, such as the deposition rate, the Ar pressure, and the substrate temperature. Kim and Oliveria
have theoretically showed that $H_C$ is inversely proportional to $t_{FM}$ for Bloch wall motion and proportional to $t_{FM}$ for Néel wall [11]. According to the theoretical model, Soohoo suggested that for ultrathin FM layer films with Néel wall the substrate roughness and the local thickness fluctuation $d t_{FM}/dx$ might be the major reason for the decreasing of $H_C$ with increasing $t_{FM}$ [9,12], and the coercivity is proportional to $d t_{FM}/dx$. In our experiments, all parameters for Co layer films with various thickness are identical except a unique variable of the film thickness with the wedge shape. In addition to the roughness of bottom interface, the wedge shape of the FM layer introduces another contribution to fluctuation $d t_{FM}/dx$. The roughness of the top interface and the local fluctuation are expected to decrease with increasing layer thickness. This leads to a reduction in $H_C$ with increasing $t_{FM}$.

In order to analyze the mechanism of $H_C$, the angular dependence was measured. For Co and CoNi single layer films, the typical results are shown in Figs. 4 and 5, respectively. When the external magnetic field is perpendicular to the easy axis, the hysteresis loop is still squared. Moreover, for CoNi single layer films, the angular dependence can be fitted just by considering uniaxial anisotropy in all angle region except narrow regions around 90° and 270°. The angular dependence of $H_C$ shows a fourfold symmetry.

From the results of the two series of single layer films in Figs. 4(c) and 5(c), one can find that the angular dependence of the in-plane resonance field $H_{res}$ can be ascribed to the uniaxial anisotropy, which is induced by the deposition process.
The uniaxial anisotropic field $H_K$ can be fitted. As shown in Fig. 3, for Co single layer films $H_K$ does not change monotonically with $t_{FM}$ and has a minimum at $t_{FM}$ of 40 nm, which is located near the center of the wedge sample. This is because $H_K$ is determined by the magnitude of the deposition field and in our sputtering system the latter one is not uniform along the wedge direction with a minimum near the center of the sample. For Co and CoNi single layer films, the angular dependence of $H_C$ does not agree with that of $H_K$. Moreover, $H_C$, i.e., critical field for the motion of the domain wall is larger than the uniaxial anisotropy. Therefore, $H_C$ cannot be described by the uniaxial anisotropy and the magnetization reversal process cannot be described by coherent rotation model. Since the magnetization reversal process of single FM layer films has great influence on variation of the output signal in GMR and TMJ devices with stray fields from the magnetic recording media, above experimental results are of crucial importance to GMR and TMJ devices [8].

In the following part, we elucidate the characteristics of the exchange biasing for as-prepared and post-FC Co/FeMn bilayers. Fig. 6 shows the experimental results. As shown in Fig. 6(a) and (b), the angular dependence of $H_E$ and $H_C$ is similar to those of conventional FM/AFM bilayers with post-FC procedure [13,14]. The angular dependence of $H_E$ and $H_C$ can be fitted by the following equations

$$H_E = \sum_{n=\text{odd}} a_n \cos (n \theta_H)$$

(1)

$$H_C = \sum_{n=\text{even}} b_n \cos (n \theta_H)$$

(2)

where $\theta_H$ is an azimuthal angle between the external field and the unidirectional axis. $a_1 = 45$, $a_3 = -9$, $a_5 = 2$, and $b_2 = 18$, $b_4 = 16$, $b_6 = 6$, $b_8 = 5$, in the unit of osterd.

Fig. 6(c) shows typical angular dependence of $H_{res}$ for as-prepared Co/FeMn bilayers. It can be fitted just by considering the unidirectional and uniaxial anisotropies. The usual expression for resonance field can be written as follows [15,16]:

$$H_{res} = H_{ISO} - H_E(\text{FMR}) \cos \theta_H - H_K \cos 2\theta_H$$

(3)

where $H_E(\text{FMR})$ is the FMR-measured $H_E$ and the uniaxial anisotropy field $H_K = \frac{2K_{U}}{M_{FM}}$, $K_U$ and $M_{FM}$ are the uniaxial anisotropy energy and the FM magnetization, respectively. $H_{ISO}$ is taken as the average value of $H_{res}$. It was suggested to come from the irreversible rotation of the AFM spins [17,18]. $H_E(\text{FMR})$ and $H_K$ can be obtained by fitting the experimental results with Eq. (3).

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**Fig. 5.** Angular dependence of (a) remanent ratio, (b) $H_C$, and (c) $H_{res}$ at room temperature for 15 nm thick CoNi single film. The dashed lines in (a) and (c) refer to fitted results based on uniaxial anisotropy and coherent rotation model.

**Fig. 6.** Angular dependence of (a) $H_E$, (b) $H_C$, and (c) $H_{res}$ at room temperature for as-prepared Co/FeMn bilayer. The dashed lines refer to fitted results based on Eqs. (1)–(3).
Fig. 7 shows the dependence of $H_E$, $H_C$, and $H_K$ on $t_{FM}$ for as-prepared and post-FC Co/FeMn bilayers. The exchange biasing can be established by deposition field during preparation and post-FC procedure, in which the Co/FeMn bilayers were cooled from 450 K to room temperature under an external magnetic field of 10 kOe [16]. As shown in Fig. 7(a), for as-prepared and post-FC samples $H_E$ is approximately proportional to $1/t_{FM}$, demonstrating the interfacial nature of the exchange biasing. Apparently, $H_E$ of the post-FC samples is much larger than that of the as-prepared samples. According to the slope of the curve, one can calculate the exchange coupling energy. For the as-prepared and post-FC samples, the exchange coupling energy is $2 \times 10^{-5} \text{ J/m}^2$ and $5.7 \times 10^{-5} \text{ J/m}^2$, respectively. Similar phenomenon was observed in Py/FeMn bilayers by Sang et al. [5]. Moreover, the value of $H_E$ determined by VSM is little larger than that determined by FMR. Some groups showed that the FMR-measured exchange field is smaller than that by VSM measurements and others showed that they are equal to each other [19,20]. This might be due to different mechanisms of the two techniques, i.e., reversible and irreversible.

As shown in Fig. 7(b), for both as-prepared and post-FC samples, $H_C$ along the easy axis decreases with increasing $t_{FM}$ as a linear function of $1/t_{FM}$. $H_C$ for the latter ones is much larger than that of the former ones. In general, for conventional FM/AFM bilayers $H_E$ and $H_C$ are usually independent of the cooling field if it is large enough to saturate the sample [21]. Therefore, the smaller $H_E$ and $H_C$ of the as-prepared ones results from the unsaturation of the FM layers during the deposition of the AFM layers. Moreover, for the as-prepared Co/FeMn bilayers, $H_K$ is almost independent of $t_{FM}$, which is controversial to the interfacial nature of the exchange biasing and $H_K$ is much smaller than $H_C$. Therefore, for the as-prepared ones the characteristic of the $H_C$ cannot be attributed to the so-called anisotropy model [22].

Figs. 3 and 7 show that $H_C$ of Co single layer films and Co/FeMn bilayers is a linear function of $1/t_{FM}$. The $H_C$ enhancement should also be a linear function of $1/t_{FM}$, as shown in Fig. 7(c). Since for single layer films and bilayers, the microstructures of the Co layers and at Co/Ag bottom interface are the same, the $H_C$ enhancement is only induced by the AFM layers. The proportional dependence clearly demonstrates the interfacial nature of the exchange biasing. In order to accurately obtain the $H_C$ enhancement induced by exchange biasing, $H_C$ of corresponding free FM layers must be removed if $H_C$ of single layer films is comparable to that of bilayers. This is true for the as-prepared Co/FeMn bilayers. If $H_C$ of the free FM films is negligible in comparison with that of bilayers, the $H_C$ enhancement is almost equal to that of the bilayers.

From the angular dependence of $H_{res}$ for the Co single layer films and as-prepared Co/FeMn bilayers, one can obtain their average values of $H_{res}$. As shown in Figs. 4 and 6, one can find with the same $t_{FM}$ the average value for...
bilayers is smaller than that of single layer films. Since the AFM FeMn layers were deposited on Co layers, the Co layers have the same microstructure in the single layer films and the bilayers and the difference in the resonance field is induced by the AFM FeMn bilayers. It is defined as $H_{\text{ISO}}$, as symbol of the exchange biasing in FM/AFM bilayers. As shown in Fig. 8, the $H_{\text{ISO}}$ is negative, in agreement with those observed in FM/AFM bilayers with normal FC procedure. However, the sign is opposite to that of the wedged-permalloy/FeMn bilayers, which was assumed to be induced by interfacial diffusion [16]. Remarkably, $H_{\text{ISO}}$ is a linear function of the inverse Co layer thickness. The linear dependence of $H_{\text{ISO}}$ further indicates the interfacial nature of the exchange biasing [16].

**4. Conclusions**

Co/FeMn bilayers, and Co and CoNi single layer films were prepared by DC and RF magnetron sputtering. Exchange biasing of bilayers and magnetic properties of these single layer films are studied. For ultrathin Co and CoNi single layer films, $H_C$ at room temperature is a linear function of $1/t_{\text{FM}}$. It is suggested to come from the interfacial roughness. The angular dependence of $H_C$ shows a fourfold-like symmetry although only the uniaxial anisotropy exists. Since $H_C$ and $H_K$ have different dependence on $t_{\text{FM}}$, the magnetization reversal process cannot be described by coherent rotation model.

The exchange biasing in as-prepared Co/FeMn bilayers has the following characteristics. First, $H_E$ and $H_C$ enhancement are proportional to the inverse Co layer thickness. However, $H_E$ and $H_C$ of the as-prepared samples are much smaller than those of post-FC samples. Secondly, the angular dependence of $H_E$, $H_C$, and $H_{\text{res}}$ is in good agreement with the reported results of post-FC ones. Thirdly, negative $H_{\text{ISO}}$ is found to be a linear function of the inverse Co layer thickness. Above three features are similar to those of post-FC FM/AFM bilayers. However, for as-prepared Co/FeMn bilayers, the $H_C$ along the easy axis is much larger than the uniaxial anisotropic field. Therefore, the magnetization reversal process of as-prepared Co/FeMn bilayers cannot be described by coherent rotation model.

The present work about the as-prepared FM/AFM bilayers might be helpful to fabrications of the GMR and MTJ devices.

**Acknowledgments**

This work was supported by the National Science Foundation of China (Grant Nos. 10174014, 60271013, 60490290, and 10321003), the State Key Project of Fundamental Research (Grant No. 2002CB613504), and Shanghai Science and Technology Committee (0252nm004).

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