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# Enhancement of exchange bias with crystal orientation in NiFe/CoO and CoO/NiFe bilayers grown on MgO(100) and MgO(111)

R. Topkaya, S. Kazan *

*Department of Physics, Gebze Institute of Technology, 41400 Cayırova, Gebze, Kocaeli, Turkey*

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ABSTRACT

We present the comparative study of the structural and magnetic properties of the NiFe/CoO and CoO/NiFe bilayers grown on both MgO(111) and MgO(100) substrates by using the ion beam sputtering technique. We observed that crystallographic orientation and crystal quality strongly affected the exchange bias properties of the bilayers. The NiFe layers showed fourfold magnetocrystalline anisotropy when we used MgO(100) substrate and uniaxial anisotropy when we used MgO(111) substrate. When the CoO layer is grown on MgO(111) and MgO(100) substrates as a first layer, instead of the NiFe, the increase of the crystalline quality and stoichiometric CoO phase has been achieved so that the exchange bias field increases by five and three times for NiFe/CoO/MgO(111) and NiFe/CoO/MgO(100) systems, respectively. The blocking temperature significantly increases for NiFe/CoO on MgO(111) and (100) substrates, indicating the increase of the stoichiometric stable $Co_1O_1$ phase in the exchange-biased system. The magnitude of the exchange bias field is 800 Oe at 10 K for NiFe/CoO/MgO(111) and more than two times as compared to that of NiFe/CoO/MgO(100). This high exchange bias value observed for NiFe/CoO/MgO(111) was explained with the uncompensated spins in CoO(111) surface. We also carried out training effect measurements to observe the durability of the exchange bias for technological applications.





## 1. Introduction

The discovery of a new type of anisotropy (exchange bias) has contributed to spin based electronic devices such as spin valves, magnetic tunnel junctions (MTJs) used for magnetic random access memory (MRAM) and magnetic field sensors [1–3]. The exchange bias (EB) originating from an interfacial exchange coupling between a ferromagnetic (FM) and an antiferromagnetic (AFM) material gives rise to the shift of the hysteresis loop of the FM material in the field axis by the amount of the exchange bias field ($H_{EB}$) after it is cooled under an external magnetic field below the Néel temperature of the AFM material.

Various theoretical models have been proposed in order to elucidate the mechanism of the exchange bias effect, most of which admit the presence of ferromagnetic, antiferromagnetic or even biquadratic interface coupling between FM and AFM layers. In the classic simple Meiklejohn-Bean model [4,5], the unidirectional anisotropy resulting from the exchange coupling of the spins at interface between FM and AMF layers is introduced into the free energy expression and this interface is regarded as fully uncompensated. However, this model estimates the EB field value larger than the experimental value. The second theory (called as random field model) explains the role of the interface spins at compensated and uncompensated interfaces between FM and AFM layers in the exchange bias phenomenon [6]. To elucidate the discrepancy between the experimental and theoretical exchange bias values, Mauri et. al. [7] suggested the theory (called as "Mauri's model") based on the formation of domain walls near the FM/AFM interface during the reversal of the ferromagnetic moments. Despite of the experimental results proving Mauri's model, this type model is not applicable to ultrathin antiferromagnetic films. Since discovered by Meiklejohn and Bean [4], the EB effect has been studied in detail by many scientists. Comprehensive researches have shown that the exchange bias effect results from uncompensated interfacial spins pinned in the antiferromagnet [8,9]. The pinned spins are fixed and they do not rotate with the applied field, and are the origin of exchange bias loop shift. The unpinned (or rotatable spins) follow the magnetization of the ferromagnet and are the origin of the coercivity enhancement. Camarero et. al. [10] indicated that the external magnetic field influences the reorganization of net uncompensated AFM spins and thus the exchange anisotropy. The exchange bias in $Ni/FeF_2$ bilayers was observed to arise from an antiparallel coupling between the ferromagnet and uncompensated Fe spins pinned in the $FeF_2$ layer [11]. Pinned (unpinned) uncompensated AFM interfacial spins were proved to induce the exchange bias in

* Corresponding author. Tel.: +90 262 605 13 21.
*E-mail address:* kazan@gyte.edu.tr (S. Kazan).

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perpendicular exchange coupled $(Pt/Co)_n$/FeMn films [12]. However, the nature and microscopic origin of the exchange bias have not still been unveiled completely [13].

It is theoretically accepted that the exchange bias field is zero for ideally compensated interface, while it is considerably high for ideally uncompensated interface [14]. However, in the comparative studies of the exchange biased bilayers having uncompensated and compensated CoO surfaces, the contradictory results have been reported. Gokemeijer et al. [15] observed the exchange bias only for the CoO(111) orientation in Py/CoO(111) and Py/CoO (100) bilayers. Surprisingly, the exchange bias was observed for the compensated AFM surfaces in $Fe/FeF_2$ systems [16]. Van der Zaag et al. [17] did not observe the significant differences in the exchange bias for the (001) and (111) orientations in $Fe_3O_4$/CoO epitaxial bilayers. Młyńczak et al. [18] reported the exchange bias for both orientations in Fe/CoO(111) and Fe/CoO(100) bilayers. Taking into account all of these studies, it is understood that the exchange bias depends not only on the crystal orientation, but also on the crystal quality, stoichiometry and interface.

In exchange bias systems, CoO has continuously been used as an antiferromagnetic material [15,18–25] because it has easily attainable low Néel temperature close to room temperature, simple NaCl-like crystal structure and strong magnetocrystalline anisotropy. Neutron diffraction studies reveal that in bulk CoO, CoO(111) surfaces are generally uncompensated, whereas CoO (100), CoO(010) and CoO(001) surfaces are completely compensated [15,26]. Moreover, CoO(111) surface spins have ferromagnetic order despite the fact that CoO is antiferromagnetic nature in a bulk state [27]. In this respect, the strong exchange bias field is observed for an uncompensated CoO(111) surface. In this study, we have investigated in detail the crystallographic orientation dependence of the exchange bias effect in NiFe/CoO and CoO/NiFe bilayer thin films grown on both MgO(111) and MgO(100) substrates by using X-ray diffraction (XRD), ferromagnetic resonance (FMR) and vibrating sample magnetometer (VSM) techniques. In the literature, in the exchange biased bilayer systems including CoO as AFM, the observed exchange bias value is generally small [15,18,20,21,28,29]. When the CoO layer is grown on MgO(111) and (100) substrates, instead of NiFe layer, the exchange bias considerably enhances due to the relatively good lattice match between CoO and MgO. We observed that with the grown of the CoO layer on MgO(111) substrate, the highest exchange bias value (800 Oe) was obtained compared to that of all the samples. The uniaxial anisotropy was observed for CoO/NiFe/MgO(111) and NiFe/CoO/MgO(111) systems, whereas the cubic anisotropy was observed as well as uniaxial anisotropy for CoO/NiFe/MgO(100) and NiFe/CoO/MgO(100) systems at the in-plane FMR measurements. It was further observed that the blocking temperature greatly enhanced for the NiFe/CoO/MgO(111) and (100) systems. We also carried out training effect measurements for the bilayer systems to observe the durability of the exchange bias field value.

## 2. Experimental

Py/CoO and CoO/Py bilayers (Py=NiFe) were grown on MgO (111) and MgO(100) substrates by using the ion beam sputtering technique with a base pressure of $\sim 10^{-9}$ mbar. This pressure range is sufficient to grow thin films of high quality in sputtering systems. Py was deposited from an alloy target and CoO was grown by reactive sputtering. A 1 cm × 1 cm × 0.5 mm polished MgO substrate with lattice constant of 4.212 Å was used. MgO substrate was cleaned three times with ethyl alcohol before transfer into the sputtering chamber. The MgO substrates were thermally treated in the sputtering chamber to remove the organic compounds. Firstly, the MgO substrates were heated up to 500 °C for 30 min prior to film deposition under high vacuum conditions and then their temperatures were gradually decreased to room temperature with steps of 50 °C. Layer, the MgO substrate surface was bombarded with $Ar^+$ ions. After O gases were introduced into vacuum chamber up to $10^{-3}$ mbar, antiferromagnetic CoO layers were grown on both MgO(111) and MgO(100) substrates by sputtering Co in a mixture of 5 mTorr Ar and 0.4 mTorr $O_2$. Following the CoO deposition, Py layer was grown on CoO/MgO under optimal growth conditions. The prepared bilayers were covered by $Al_2O_3$ cap layer to prevent oxidation of bilayer structures. By using the similar steps, CoO/Py bilayers were grown on both MgO(100) and MgO(111) substrates. To avoid the effect of thickness on the magnetic properties of thin film, we fixed the thickness of CoO and NiFe layer in all fabricated films. The thickness of both CoO and NiFe layers and roughness at the interface were checked by small angle X-ray reflectivity measurements. The thickness of CoO and NiFe layer were determined as 4.5 (±0.1) nm and 9.6 (±0.1) nm, respectively. FMR measurements were carried out using commercial EMX X-Band (9.8 GHz) spectrometer at room temperature. The exchange bias measurements of CoO/NiFe bilayer were carried out using the Quantum Design Physical Property Measurement System (PPMS) with an applied magnetic field parallel to the film surface.

## 3. Theoretical model

The ferromagnetic resonance (FMR) was proved to be powerful and useful technique in order to determine magnetic properties of the samples [30–33]. FMR spectra were recorded for the in-plane and out-of-plane geometries. For FMR measurements at the in-plane geometry (IPG), the sample was horizontally attached to the bottom edge (horizontal flat surface) of the sample holder. The static magnetic field lies in the sample plane at this geometry. For FMR measurements at the out-of-plane geometry (OPG), the sample was vertically attached to the front of the sample holder. The magnetic field component of microwave lies in the film plane and the static magnetic field is rotated from the sample plane toward the film normal at this geometry. The experimental coordinate system, the relative orientation of the magnetization vector ($M$) and the applied static magnetic field vector ($H$) and picture of the prepared bilayer structure were illustrated in Fig. 1. In order to obtain the theoretical data for fitting to the experimental data, the magnetic energy density is assumed as

$$E = -MH + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_u \sin^2\theta + 2\pi M^2 \cos^2\theta - K_\perp \cos^2\theta \quad (1)$$

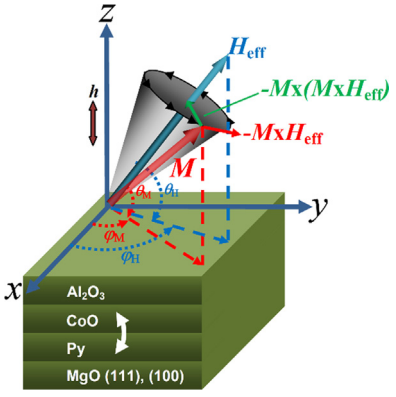


**Fig. 1.** Relative orientations of the external magnetic field ($H$) and magnetization vectors ($M$), the used coordinate system and picture of the prepared bilayer structure.



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