Tailoring coercivity of unbiased exchange-coupled ferromagnet/antiferromagnet bilayers

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This paper reports experimental results obtained on unconventional exchange-coupled ferromagnet/antiferromagnet (FM/AF) system showing zero net bias. The Curie temperature of the FM (NiCu) is lower than the blocking temperature of the AF (IrMn). Samples were either annealed or irradiated with He, Ar, or Ge ions at 40 keV. Due to the exchange coupling at the FM/AF interface, the coercivity ($H_C$) of the as-deposited FM/AF bilayer is rather higher than that of the corresponding FM single layer. We found that by choosing a proper ion fluence or annealing temperature, it is possible to controllably vary $H_C$. Ion irradiation of the FM single layer has lead to only a decrease of $H_C$ and annealing or He ion irradiation has not caused important changes at the FM/AF interface; nevertheless, a twofold increase of $H_C$ was obtained after these treatments. Even more significant enhancement of $H_C$ was attained after Ge ion irradiation and attributed to ion-implantation-induced modification of only the FM layer; damages of the FM/AF interface, on the other hand, decrease the coercivity. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4731717]

I. INTRODUCTION

In general, the most important property of systems composed of a ferromagnet (FM) exchange-coupled to an antiferromagnet (AF) is the exchange bias (EB) responsible for the shift of the static magnetization curve along the magnetic field axis. The EB is frequently accompanied by an increase of the coercivity ($H_C$), i.e., the half width of the magnetization hysteresis loop. This enhancement of $H_C$ in EB systems seems promising for applications in information storage devices. Formerly, efforts to manufacture permanent magnets with improved properties have been focused on producing the so-called exchange-spring magnets where FM/FM exchange coupling between hard and soft materials is utilized. The potential of FM/AF systems for applications in high-energy-product permanent magnets has usually been studied in FM transition-metal particles coated with their native AF oxide or native sulfides and nitrides, in mixtures of mechanically alloyed components, as well as in phase-separated alloys. In MnF$_2$/Fe and FeMn/NiFe bilayers, the increase of $H_C$ has been ascribed to spin pinning at the FM/AF interface.

Normally, the magnetic properties on EB systems are manipulated by means of magnetic anneal and, most recently, via ion irradiation/implantation. The latter has been shown to be a useful tool for modification of the structural and compositional properties of magnetic materials through the so-called magnetic patterning. Chemical phase transition could be induced via ion irradiation and this can even lead to ferromagnetism in naturally paramagnetic systems. In EB systems, both EB field magnitude and EB direction can be modified through ion bombardment. EB enhancement can be achieved for some systems in a certain fluence range if the ion bombardment is performed in the presence of magnetic field.

Here, we present results on an unconventional magnetron-sputtered IrMn/NiCu system which does not present EB at room temperature (RT). Pieces of the as-made IrMn/NiCu bilayer and of a NiCu single layer were irradiated with He, Ar, or Ge ions at different fluences or annealed at different temperatures ($T_{ANN}$). The unconventionality of our exchange-coupled FM/AF system refers to the fact that the Curie temperature ($T_C$) of the FM layer, Ni$_{75}$Cu$_{25}$, is rather lower than the Néel temperature ($T_N$) of the IrMn AF layer. One notes that despite a number of very interesting effects related to FM/AF coupling observed in thin films and in AF nanoparticles with ferromagnetic shells with $T_N$ higher or close to $T_C$ and regardless of their potential technological applicability, studies on such systems are still rather scarce owing to the difficulties in initializing and manipulating the EB effect.

We show that despite our system does not present EB, owing to the exchange coupling at the FM/AF interface the coercivity of the FM/AF bilayer is rather higher than that of the FM single layer in the as-made state. Furthermore, we demonstrate that $H_C$ can be controllably tuned by means of post-deposition annealing or ion irradiation.

II. EXPERIMENTAL

IrMn/NiCu bilayer was grown at RT onto naturally oxidized Si(100) substrate in a magnetron-sputtering AJA ORION 8 UHV system at base pressure of $1 \times 10^{-7}$ Torr. The 7 nm thick IrMn layer was DC-sputtered from an Ir$_{30}$Mn$_{70}$ target in a 7.5 mTorr Ar atmosphere at a deposition rate of 1.13 Å/s. The subsequently grown Ni$_{25}$Cu$_{25}$ (30 nm) layer was co-sputtered from Cu and Ni targets, both in a 2 mTorr Ar atmosphere. The Ni and Cu deposition rates (0.98 and 0.37 Å/s, respectively) were suitable for producing a NiCu layer with low $T_C$; also, $T_C$ was intended to be around 373 K which is the high-temperature limit of our measurements facilities. A Ni$_{15}$Co$_{85}$ (6 nm) buffer layer was obtained by co-deposition in order to promote IrMn(111) texture, and a 4 nm thick Cr layer was grown on top to protect the FM.
layer from oxidation. The advantage of this composition of NiCr is that it is non-magnetic. A film containing only an FM layer, i.e., Si(100)/Ni75Cu25(30 nm), was also deposited. A static magnetic field of 130 Oe was applied in the plane of the films during deposition.

The structural characterization, before and after the post-deposition treatments, was carried out by means of conventional x-ray diffractometry (XRD) using Philips X’Pert MRD machine employing Cu Kα radiation. Thermomagnetic curves were obtained on the FM/AF system using a superconducting quantum interference device. The magnetic properties of the as-made films were modified by either irradiation with He, Ar, or Ge ions at different fluences or by thermal annealing. The RT magnetic characterization of the samples was made via static magnetization (M) curves using an alternating gradient-force magnetometer with magnetic field, H, applied in the plane of the films.

Ion irradiation was performed using 40 keV He⁺ or Ar⁺ beams at a current density of 100 nA/cm² and at fluences ranging from 3 × 10¹³ to 1 × 10¹⁵ ions/cm². Also, a 40 keV Ge⁺ beams at a current density of 50 nA/cm² and fluences from 5 × 10¹⁵ to 1 × 10¹⁵ ions/cm² were used. The irradiations were performed in a presence of a 5.5 kOe in-plane magnetic field parallel to that applied during the sputtering.

Pieces of both FM/AF bilayer and FM single layer were also heated in an electric resistive furnace in vacuum (pressure better than 10⁻⁶ mbar), kept for 5 min at TANN ranging between 360 and 530 K, and then cooled down to RT, always upon a magnetic field of 3.6 kOe, also parallel to the direction of the field applied during the deposition.

III. RESULTS AND DISCUSSION

T_C of the FM layer of the IrMn/NiCu film was determined from a pair of zero-field-cooled/field-cooled magnetization versus temperature (T) curves (not shown), being the field-cooled one measured upon a 100 Oe magnetic field; T_C, defined as T that corresponds to the maximum slope of M(T), was estimated to be 353 K. This value is lower than both the blocking temperature of Ir20Mn80 (≈520 K) and the respective T_N (≈680 K).

The top panels of Fig. 1 give the depth profiles of the ion distributions into the IrMn/NiCu sample after 40 keV He⁺ or Ge⁺ irradiations calculated using SRIM simulation. It is seen that while practically all He ions pass through the deposited layers, the great majority of Ge ions are implanted into the NiCu layer. X-ray diffraction patterns obtained at RT on pieces of the IrMn/NiCu bilayer before (as-deposited film) and after He⁺ (using fluence of 5 × 10¹⁵ ions/cm²) or Ge⁺ (1 × 10¹⁴ ions/cm²) irradiation are plotted in the bottom panel of Fig. 1. Apparently, both IrMn and NiCu layers are polycrystalline exhibiting (111) texture. Given that the three x-ray diffraction patterns practically coincide, this analysis does not indicate important changes in the crystalline structure of the FM/AF bilayer after any of these post-deposition treatments. There is a possibility of forming a Ni-Ge phase as a result of the Ge⁺ implantation into the NiCu layers. In NiFe thin films, Ge incorporation can influence on the mode of the magnetization reversal. Ge concentration higher than 1% could lower T_C of Ge implanted Ni-Ge alloys; however, since the SRIM simulations gave Ge concentration lower than 0.05% in our FM films, the influence of such a phase (if exists) on the NiCu magnetic properties should be insignificant.

Although none of our samples showed EB, when pieces of the as-made FM/AF film were annealed or irradiated with either He⁺ or Ge⁺, significant changes in their magnetic behavior were observed. The top panel of Fig. 2 shows representative M(H) obtained on pieces of the bilayer annealed at three different temperatures; the loop that corresponds to the as-made sample is also given for comparison. Magnetization curves traced after He⁺ or Ge⁺ irradiation for three representative fluence values are given in the middle and bottom panels of Fig. 2, respectively. No significant modification of the remnant magnetization, M(H = 0), after any of the treatments is observed.

The coercivity, however, changes considerably. The dependencies of H_C on TANN and on the He⁺ or Ge⁺ fluence are given in Fig. 3. Each of these variations, relative to the FM/AF bilayer, shows a maximum. H_C of the anneal series presents a broad peak at approximately 420 K, where the coercivity value is almost twice higher than that of the untreated film. The variation of H_C after He⁺ irradiation, however, shows a rather sharp cusp corresponding to a fluence of 5 × 10¹³ ions/cm² where H_C is twice higher than the as-made sample value, followed by a gradual decrease with the fluence, returning to the initial value for the highest fluence of 1 × 10¹⁵ ions/cm². Most important is the coercivity enhancement after implantation with Ge⁺ employing a fluence of 1 × 10¹⁴ ions/cm² when H_C = 42 Oe is measured, i.e., an approximately threefold enhancement is achieved. For higher Ge⁺ fluences, H_C decreases very rapidly at first and, for the highest fluence used, reaches one fifth of the value of the as-made bilayer.

The empty symbols in Fig. 3 represent the H_C data obtained after ion irradiation of the FM single layer sample.
The respective hysteresis loop traced before any post-deposition treatment is plotted in the top panel of Fig. 4 where, for comparison, the loop of the as-made FM/AF bilayer is also given. The curves that correspond to the FM single layer after He\(^+\) or Ge\(^+\) irradiation are given in the bottom panel of this figure. Contrary to that obtained for the FM/AF bilayer, these treatments lead to reduction of the coercivity only. The hysteresis loops traced on the FM/AF bilayer after the Ar\(^+\) irradiations (not shown) are very similar to those of the irradiated FM single layer, presenting practically the same decrease of \(H_C\) as well, i.e., only a monotonic decrease of the coercivity with the Ar fluence is observed.

From these experimental results, it becomes clear that significant variations in \(H_C\) are only attained in the FM/AF bilayer, which shows rather higher coercivity than the FM single layer even in the as-made state. Enhancement of \(H_C\) due to exchange coupling between a FM layer and AF interface layer magnetization, irrespective of whether the AF is structurally disordered or not, has been estimated by model calculations, see, e.g., Refs. 25 and 26. In most cases, changing \(H_C\) of an FM requires variations of the density or nature of the defects in the sample, its crystallinity or domain size. As the x-ray diffraction analysis indicates, changes of the crystallographic structure of the FM layer should be discarded as an essential source for the coercivity enhancement of our FM/AF films. This statement is also sustained by the non-observation of such an effect in the respectively-treated FM single layer.

Therefore, the main cause for the modification of \(H_C\) in the IrMn/NiCu films seems to be different from that responsible for the gradual coercivity decrease in conventional FM/AF bilayers with \(T_C \gg T_N\). It has been recently shown that point defects (interstitial and vacancies) in the bulk part of IrMn layers, produced by ion bombardment using H, He, and Ne ions at different fluences and currents, lead to a decrease in \(H_C\) independently on other ion-irradiation effects as long as the sample is not too damaged.\(^{27,28}\) Magnetic frustration at the AF part of the interface due to annealing upon application of magnetic field with proper strength has been pointed out by Leighton et al.\(^{10}\) as responsible for the strong enhancement of \(H_C\) in MnF\(_2\)/Fe bilayers. They have found that when the AF surface is in a state of maximum magnetic frustration and zero net bias, this effect is proportional to the exchange coupling between the AF and FM layers. According to these authors, frustrated FM/AF interfaces provide local energy minima which effectively pin the propagating domain walls in the FM. In other words, the AF surface splits similarly to those of the irradiated FM single layer, presenting practically the same decrease of \(H_C\) as well, i.e., only a monotonic decrease of the coercivity with the Ar fluence is observed.

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into magnetic regions which are aligned either with the magnetic field or are in the original AF-coupled configuration. However, seeing as our films do not present noteworthy anisotropy induced by the field applied during the treatments, we can disregard this mechanism as well.

The absence of preferential magnetization orientations of both FM single layer and FM/AF systems could be explained as follows. Due to its polycrystalline structure, the as-made FM single-layered film with very low intrinsic anisotropy constant is magnetically isotropic in-plane which results in insignificant $H_C$. The low $M_s$ and $T_C$ values of NiCu do not favor an easy axis to be induced either during the film’s growth or through any of the post-deposition treatments. The noteworthy FM/AF bilayer’s coercivity clearly indicates that its magnetic anisotropy, resulting in appreciable $H_C$, is solely due to the presence of the IrMn layer. The very strong anisotropy of the latter and the (apparently) weak NiCu/IrMn exchange coupling do not tolerate reorientation of the spins of the IrMn sublayer adjacent to the NiCu one, even in the state of NiCu saturation. The AF layer is polycrystalline and the topmost IrMn sublayer is magnetically compensated and with random in-plane spins’ orientations before annealing or ion irradiation. Since it resides like that after any post-deposition treatment, no easy axis or EB is observed.

There are two other mechanisms that can lead to the enhancement of $H_C$ of our unbiased FM/AF system:

1. Variations of the grain size of the AF after magnetic anneal or ion irradiation can be responsible for the raise of the coercivity. Li and Zhang, via model calculations, have shown that the enhanced coercivity in FM/AF bilayers can be explicitly related to experimentally controllable parameters such as the FM layer’s thickness and the grain size of the AF. These authors have obtained that when the interface FM/AF interaction is stronger than the coupling into the FM layer, the AF layer breaks the adjacent FM into domains with size smaller than that of the FM layer alone, which break-up is essential for the increased coercivity.

2. Post-deposition-induced increase of the interfacial roughness could raise the interfacial area and might influence the interfacial coupling by causing local spin dispersion resulting in additional local anisotropies. These anisotropies provide an energy barrier to domain-wall motion thus increasing the coercivity. Higher-order anisotropy contributions arising from the exchange interaction between AF and FM layers have also been found and believed to have a strong influence on the coercivity.

The above two mechanisms, acting separately or together, should be considered as responsible for the coercivity enhancement of our FM/AF bilayer. However, the causes for the distinct types of variation of $H_C$ with the fluence for the cases of light-ion (He$^+$) irradiation, or implantations with rather heavier ions (Ar$^+$ and Ge$^+$) are different. Since the electronic stopping power predominates for 40 keV He$^+$ irradiation, its main effect is local hyperthermal heating triggered by electronic excitations. This is confirmed by Fig. 5 where the estimated via SRIM simulations depth profiles of the damages caused by 40 keV He$^+$, Ar$^+$, or Ge$^+$ irradiations into Si/NiCr/IrMn/NiCu/Cr films are shown. It is seen that He$^+$ irradiation does not cause important damages at any of the deposited layers thus explaining the practically equal maximum values of $H_C$ attained either after annealing or He$^+$ irradiation as well as the fact that neither of these two treatments leads to a decrease of $H_C$ as compared to that of the as-made FM/AF bilayer. The trend of minor coercivity increase for high temperature anneals, see the top panel of Fig. 3, should mainly be attributed to an improvement in the bulk IrMn crystallinity and/or grain growth observed after a short-duration annealing.33 Seeing as the main effect of He$^+$ bombardment is hyperthermal heating which does not cause an IrMn improvement, no tendency of coercivity increase with the fluence is observed for high fluences. Most likely, the sharp peak in this fluence dependence of $H_C$ corresponds to the optima, for the particular sample and for the energy used of this room-temperature treatment, magnetic frustration and/or domain size at the AF part of the interface.

As already discussed, Fig. 1 shows that although the great majority of the Ge ions are implanted into the FM layer, important changes in the crystalline structure of the FM/AF bilayer after Ge$^+$ implantation were not detected. Figure 5 clearly illustrates that 40 keV Ge$^+$ implantation does not virtually cause damages at the FM/AF interface. Recall that the presence of AF is responsible for the higher value of $H_C$ of the as-made FM/AF film as compared to the FM single layer. Therefore, the coercivity variations with the ion fluence for the case of Ge$^+$ implantation must be predominantly attributed to changes of the magnetic structure (e.g., domain size) of the FM part of the FM/AF bilayer only. The optima conditions, for the ion energy used here, are achieved for a fluence of $1 \times 10^{14}$ ions/cm$^2$, which corresponds to the noteworthy (threefold) enhancement in $H_C$. Ge$^+$ irradiation at high fluences leads to low coercivity values very close to those measured for the FM single layer (see the bottom panel of Fig. 3). Although alike changes of the magnetic structure of the FM single layer certainly take place as a result of Ge$^+$ implantation, enhancement of $H_C$ is not observed since, as already emphasized, for this to happen exchange coupling with AF is essential.

An important question that arises from the above considerations is what would $H_C$ value be if the energy of the Ge
ion beam were increased sufficiently, for Ge+ implantation at the FM/AF interface, keeping fixed the value of the optimum (for the $H_C$ enhancement) fluence. One can answer this question using the damage depth profile relative to the IrMn/NiCu film after 40 keV Ar+ implantation plotted in Fig. 5. The Ar ions, being lighter than the Ge ones, penetrate deeper into the sample causing damages not only into the FM layer but also in the AF one and at the FM/AF interface, resulting in the observed drop of $H_C$ for Ar ion implantation as mentioned above. Most probably, the increased number of defects at the interface leads to a decrease of the AF domain size making the AF grains more unstable magnetically.

To conclude, we showed that, by choosing adequate ion fluence or $T_{\text{ANN}}$, one can obtain a controlled variation of more than one order of magnitude of the coercivity of unconventional exchange-coupled though unbiased IrMn/NiCu films. The main findings regarding the coercivity enhancement may be summarized as follows: (1) Due to the exchange coupling at the FM/AF interface, $H_C$ of the FM/AF bilayer is rather higher than that of the FM single-layered film already in as-made state. (2) Ion irradiation of the FM single layer leads to a decrease of the coercivity only. (3) Annealing or He ion irradiation does not cause important changes at the FM/AF interface. The respective twofold increase of $H_C$ attained after either of these treatments might be attributed to an improvement in the crystallinity and/or grain size of the magnetic layers. (4) The even more significant enhancement of $H_C$ after 40 keV Ge ion irradiation, observed for certain fluence values, is due to implantation-induced modification of the FM layer alone, while damages of the FM/AF interface result in changes of the AF domain size and a coercivity decrease. (6) Magnetic field applied during annealing or ion irradiation, regardless the ion, does not influence the magnetic properties of the films. It seems clear that this behavior warrants further study.

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