Mechanism of Exchange Bias Formation in (110) Al_2O_3/Py/Mn/Ta Nanostructures

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ABSTRACT

The mechanism of unidirectional exchange anisotropy at thermomagnetic treatment of permalloy - manganese bilayers has been studied. It has been found that beginning from the annealing temperature of 230^0C a hysteresis loop shift appears. The maximal exchange bias of 155 Oe is reached after the annealing at 250^0C for 2 hours. It is demonstrated by transmission electron microscopy that the exchange bias arises and the coercive force increases due to the formation of antiferromagnetic NiFeMn phase resulting from diffusion interaction of permalloy and manganese at annealing.

Keywords: exchange bias, bilayers, thermomagnetic treatment, permalloy, manganese

1 INTRODUCTION

Since the discovery of unidirectional exchange anisotropy this phenomenon has become the basis for a number of important practical applications, especially in information storage devices [1,2]. Thus, for instance, this phenomenon, also referred to as exchange bias, is widely used in spin valves which are systems consisting in the simplest case of two ferromagnetic (FM) layers separated by a non-magnetic layer and an antiferromagnetic (AFM) layer. The hysteresis loop of FM-AFM bilayers is shifted along the magnetic field axis, and its shift relative to zero is characterized by an exchange bias field (H_{ex}).

The necessary requirements for spin valves application are high exchange bias field, low relationship $H_c/H_{ex} < 1$ (where H_c is a coercive force or coercivity) and as high as possible corrosion resistance and blocking temperature, T_b, i.e. the temperature at which the exchange bias field of FM-AFM bilayer reduces to zero.

Among a great number of various exchange biasing materials, exchange couples consisting of thin films of permalloy and different antiferromagnetic films were thoroughly studied, because of a relatively high exchange anisotropy discovered in such systems [3-7].

It is known that at annealing of permalloy-manganese bilayers an ordered antiferromagnetic NiFeMn compound may form [5-7]. In such bilayers the blocking temperature after the special thermomagnetic treatment may be as high as 330^0C [3], which makes them quite promising for spin valve applications. However, conditions and mechanisms of this phase formation are not adequately understood by now.

Permalloy-manganese bilayers were first studied in [3,4], and a unidirectional anisotropy was observed at annealing beginning from 300^0C. The authors attributed the unidirectional anisotropy to the formation of a continuous antiferromagnetic NiFeMn layer at an interface between permalloy and manganese.

In later publication [5] the unidirectional exchange anisotropy in permalloy-manganese bilayers was observed after annealing in magnetic field at 240^0C, and the maximal exchange bias field was 290 Oe after the annealing at 300^0C for 4 hours. The ratio H_c/H_{ex} = 0.9 was high enough. The authors of this work also attributed the unidirectional anisotropy appearance to the NiFeMn layer formation at interlayer boundary. At 400^0C annealing of $(Ni_{80}Fe_{20})_{1-x}Mn_x$ films, where $0.05 < x < 0.6$, the NiFeMn phase was not observed, but, on the contrary, a homogeneous film decomposed into permalloy and manganese [8].

An essentially other mechanism of the unidirectional anisotropy appearance was suggested in [7]. According to the data of this study exchange anisotropy arises because of diffusion of manganese along permalloy grain boundaries and the formation of an ordered antiferromagnetic phase enveloping the grains. In this study the coercive force H_c was always higher than the exchange bias field H_{ex} independently on the thermomagnetic treatment regime.

It should be noted that in all the available publications the exchange bias field H_{ex} does not exceed 30 Oe and is of the same order of magnitude as the coercive force.

The main goal of the present study was to investigate an effect of thermomagnetic treatment on the magnetic properties of permalloy/manganese bilayers and to reveal regularities and possible mechanisms of the unidirectional exchange anisotropy formation in the bilayers under study. This system is of practical interest because it enables to ensure magnetic properties required for realization of unidirectional exchange anisotropy by an appropriate choice of the annealing regimes.

2 EXPERIMENT

The study was carried out on two-layered $Ni_{81}Fe_{19}$/Mn films deposited on single crystal Al_2O_3 (110) substrates. They were fabricated as follows. At first the $Ni_{81}Fe_{19}$ layer was deposited by ion-plasmic technique on a modernized magnetron device URM3-013 at the pressure of actuating gas (argon) of 10^{-3} Torr. The film thickness was controlled by a mechanical profilometer Dektak 150, and it was 30 nm. The as-obtained specimens were put into an electron-beam chamber Varian, preliminary heated and held at 250^0C for 1 h at the pressure of 10^{-6} Torr for the removal of atmospheric gases which could be absorbed from the air, and then manganese was sputtered on their surface. As demonstrated by Auger-electron spectroscopy, there were no gas constituents from the air both at permalloy-manganese interfaces and in the films bulk. To prevent oxidation of as-obtained films, they were covered with a thin Ta layer. The thicknesses of Mn and Ta layers were controlled by a quartz thickness indicator. The multilayer structure obtained by this procedure is as follows: $Al_2O_3/Ni_{81}Fe_{19}$(30nm)/Mn(100nm)/Ta(10nm).

The specimens under study were annealed in the vacuum of 10^{-6} Torr in a constant magnetic field of 2 kOe applied in a film plane. The hysteresis loops were measured under the same magnetic field direction as at annealing. Magnetic properties were studied in a vibrating sample magnetometer at room temperature in the range of magnetic fields of ± 2 kOe. Coercivity H_c and exchange bias field H_{ex} were determined from hysteresis loops as a loop half-width and its center shift relative to the zero coordinate along the magnetic field axis, respectively.

The films composition was studied by Auger-electron microscopy in Varian Auger spectrometer. Concentration profiles of chemical elements constituting the specimens under study were studied by ion etching, and quantitative estimation was done by means of element sensitivity coefficients.

The films structure was studied by transmission electron microscopy (TEM) on Phillips CM-30 microscope. To obtain samples for TEM studies, the substrate was carefully chopped off at a certain angle in such a manner, that a piece of film projected from it and could be examined in a normal to its plane direction.

3 RESULTS AND DISCUSSION

According to the data of electron diffraction analysis, an ordered antiferromagnetic phase NiFeMn is formed in permalloy-manganese bilayers at annealing in the temperature range of 230-250^0C. This phase formation is evidenced by electron diffraction patterns in which superstructure reflections appear when the specimens are annealed in this temperature range (Fig. 1). At lower annealing temperature the appearance of this phase is not revealed. According to TEM data no other phases are formed at thermomagnetic treatment in the temperature range studied.

The AFM phase formation is in agreement with the data of magnetic measurements.

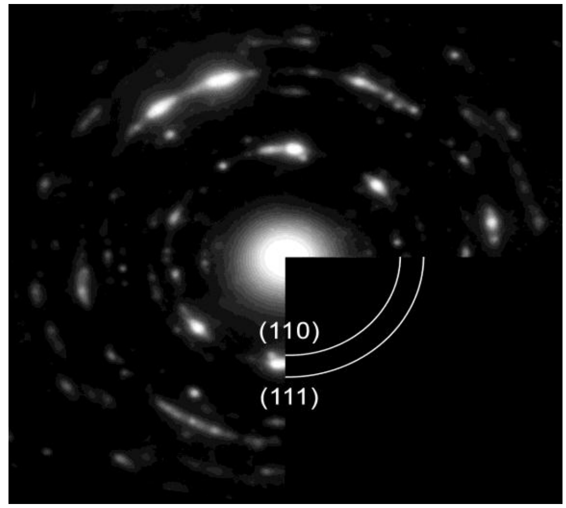

Figure 1: Electron diffraction pattern of a specimen annealed at 250 ^{0}C, 2 h

Figure 2 demonstrates hysteresis loops in the specimens directly after sputtering and after the annealing at 200^0C for 8 h. In the ordinate axis a ratio of magnetization of a specimen under study (M) to the saturation magnetization of an initial (not annealed) specimen (M_s^0) is shown. The hysteresis loop of the as-sputtered specimen is symmetrical relative to coordinate origin and its coercivity is about 1 Oe which is characteristic of soft magnetic permalloy. At 200^0C annealing the coercive force increases slightly. After 8 h annealing at this temperature it grows from 1 to 4 Oe.

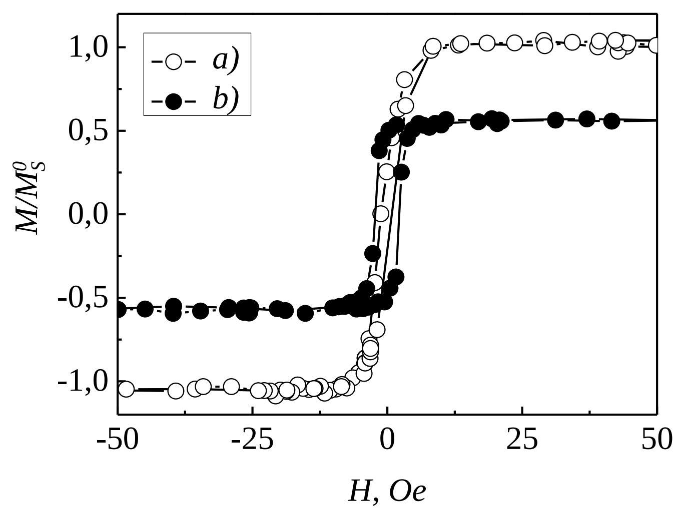

Figure 2: $Al_2O_3/Ni_{80}Fe_{20}$(30nm)/Mn(100nm)/Ta(10nm) hysteresis loops after sputtering (a) and annealing at 200^0C for 8 h (b)

The unidirectional anisotropy effect does not appear after annealing at 200^0C, because, as mentioned above, the ordered antiferromagnetic phase is not formed. The main

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effect of annealing at this temperature consists in considerable decrease of saturation magnetization which testifies the reduction of the ferromagnetic phase volume fraction. After 200^0C, 8 h annealing about 55 % of permalloy is retained in ferromagnetic state whereas the rest of this phase becomes paramagnetic.

According to the magnetic state diagram of Ni-Fe-Mn alloys [9], the $(Ni_{0.80}Fe_{0.20})_{1-x}Mn_x$ alloy at room temperature and in the composition range corresponding to x = 0.35–0.50 is in the paramagnetic state. Thus, it may be concluded from the data obtained that at this annealing temperature (200^0C) there are no areas in permalloy in which concentration of manganese is higher than 50 %.

When the annealing temperature is raised up to 230^0C, the magnetic properties behavior appreciably changes.

Figure 3 demonstrates dependences of the exchange bias field (H_{ex}) and coercivity (H_c) on the time of annealing at 230^0C. After 1 hour annealing only slight increasing of the hysteresis loop width is observed. The unidirectional exchange anisotropy after such heat treatment is absent. Consequently, it may be concluded that the 230^0C, 1 h annealing does not result in the antiferromagnetic phase formation.

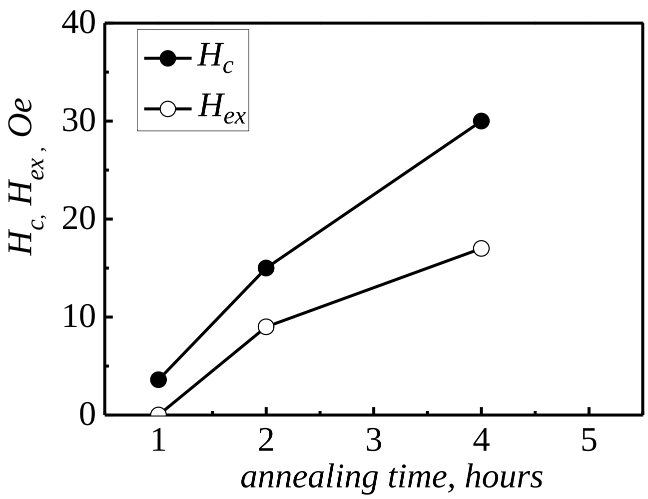

Figure 3: Dependences of exchange bias field H_{ex} and coercivity H_c of the specimen $Al_2O_3/Ni_{80}Fe_{20}$(30nm)/Mn(100nm)/Ta(10nm) on the annealing time at 230^0C.

When the annealing at 230^0C is prolonged up to 2 hours, the hysteresis loop parameters considerably change which indicates an appearance of the antiferromagnetic phase. A moderate exchange bias field of 9 Oe appears and coercivity grows up to 15 Oe. When the annealing time is increased to 4 hours, the exchange bias field increases further up to 17 Oe and the coercive force grows up to 30 Oe.

At higher annealing temperature of 250^0C the films behavior changes more rapidly (Fig. 4). Even after 1 h annealing at this temperature the exchange bias field is as high as 28 Oe and coercivity is 19 Oe. After the heat treatment for 2 h H_{ex} grows up to 155 Oe whereas the coercive force is 63 Oe. After the 4 h annealing at this temperature the magnetic moment of the specimen vanishes, indicating complete disappearance of the ferromagnetic phase in it. In this case only antiferromagnetic and paramagnetic phases are present in the multilayer.

Note that at this annealing temperature, 250^0C, the exchange bias field is higher than the coercive force. At 2 h annealing the H_c/H_{ex} ratio is minimal and equals to 0.4.

Consequently, the ordered antiferromagnetic phase NiFeMn may be considered as one of the constituents of multilayers with unidirectional exchange anisotropy. Hence, such multilayers may be promising for application in spin valves due to high blocking temperature and high values of the exchange bias fields. Based on this system spin valves with enhanced thermal stability and improved hysteresis characteristics can be fabricated.

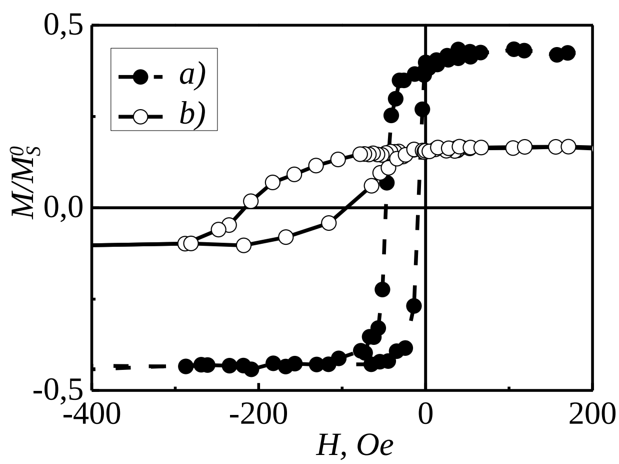

Figure 4: Hysteresis loops of the specimens $Al_2O_3/Ni_{80}Fe_{20}$(30nm)/Mn(100nm)/Ta(10nm) annealed at 250^0C for 1 h (a) and 2 h (b)

The Auger spectroscopy studies show that there are no intermediate layers at an interface between permalloy and manganese. With increasing temperature and time of annealing the ferromagnetic permalloy layer is saturated with manganese. However, even in a specimen annealed at 250^0C for 4 hours, in which the ferromagnetic phase was entirely absent, the mean concentration of Mn in permalloy layer reached only about 50 at. %.

The data obtained may be interpreted as follows. At annealing Mn penetrates into permalloy diffusing mainly along crystallite boundaries in nanocrystalline permalloy layer. According to TEM data, crystallite sizes in permalloy fall in the range of 10-20 nm. That is why the relative fraction of crystallite boundaries which are the fast diffusion paths is high enough. Besides, Mn diffusion along dislocations, low-angle boundaries and other defects is also possible. As for the volume diffusion, it is impossible at the annealing temperatures applied in this study. The latter conclusion may be done based on the available data on the volume diffusion coefficients. Unfortunately, there are no data on Mn diffusion in permalloy, and we have to rely on the available data, for instance, on the description of impurity diffusion of Cr and Fe in nickel [10]. According to Mehrer's data, the volume diffusion coefficients of Cr and Fe in Ni at 250^0C are as low as $6.5\cdot10^{-32}$ m/s and $1.2\cdot10^{-31}$ m/s, respectively. Consequently, the penetration depth in case of 4 h annealing is only about 10^{-14} m, i.e. manganese penetration into the adjacent permalloy layer by the volume diffusion mechanism is improbable. On the other hand, the penetration of this element into permalloy crystallite bulk is evidenced by the magnetic behavior. As demonstrated above, the saturation magnetization of the multilayers decreases at annealing which indicates the reduction of the ferromagnetic phase fraction. That is why we conclude that Mn penetration is realized by the fast diffusion paths such as grain boundaries, subgrain boundaries, dislocations, etc. It is the Mn diffusion into permalloy that is responsible for the saturation magnetization drop and the coercivity growth observed at annealing. Beginning from the annealing temperature of 230^0C, an ordered antiferromagnetic phase appears in the structure of the multilayers under study, and its formation results in the coercive force growth and the hysteresis loop shift. Taking into account the above consideration it is natural to assume that this phase is forming first of all at crystallite boundaries and then grows into the crystallite bulk. This mechanism is in agreement with the data obtained in [7].

4 CONCLUSION

It has been found that at annealing of permalloy-manganese bilayers in a magnetic field their coercive force increases and hysteresis loop shifts along the magnetic field axis. With increasing annealing temperature and time the ferromagnetic permalloy layer is saturated with manganese which results in the saturation magnetization decrease indicating the reduction of the ferromagnetic phase fraction. At the same time the coercivity growth is observed. Beginning from the annealing temperature of 230^0C, the heat treatment results in the formation of an ordered antiferromagnetic phase, which is responsible for the unidirectional anisotropy appearance. The maximal hysteresis loop shift observed in the present study is 155 Oe after the bilayers annealing at 250^0C for 2 hours. In this case the exchange bias considerably exceeds the coercive force.

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