Uniaxial in-plane magnetic anisotropy and exchange bias in Sm/Fe bilayers

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We have studied structural and magnetic properties of quartz/Sm(25 nm)/Fe(15 nm) thin films by grazing incidence x-ray diffraction, atomic force microscopy, longitudinal magneto-optic Kerr effect, and superconducting quantum interference device magnetometry. Both Sm and Fe layers exhibit growth in a preferential direction with textured granular structure. The Fe layer is composed of small elongated grains (36 nm long and 21 nm wide). The Fe grains are monodomains and are all parallel aligned due to the texture effect. Therefore, their magnetic shape anisotropy is summed, producing a final uniaxial in-plane magnetic anisotropy in the film. The Sm/Fe system is an antiferromagnetic/ferromagnetic bilayer exhibiting exchange bias coupling below 40 K. The long-range character of Sm-Fe spin interaction located near the interface maintains this coupling, in spite of disorder effects at the interface, such as Sm-Fe alloying, and results in a power-law decay with temperature for the exchange bias field (H_E). [S0163-1829(99)09025-6]

Over the last several years the growth of epitaxial magnetic thin films on single crystal substrates has allowed the study of a wide variety of magnetic properties.[1] Interesting results have been obtained in magnetic thin films based on rare-earth (RE) metals going from pure metal films[2] and bi- and multilayers[3] to superlattices,[4] the study of magnetic ordering in the layers as well as the magnetic coupling between them being one of the most investigated subjects.

The interlayer exchange coupling may be of long or short range, leading to various magnetic properties. The interlayer coupling through long-range interaction is well depicted in ferromagnetic layers separated by a paramagnetic one, where the exchange coupling between the ferromagnetic layers is an oscillatory and decreasing function of the paramagnetic layer thickness, and which couples the layer's moment parallel or antiparallel (for a recent review see Ref. 5). This coupling is mediated through the Ruderman-Kittel-Kasuya-Yosida (RKKY) long-range interaction.

On the other hand, exchange coupling of short range occurs in antiferromagnetic (AF)/ferromagnetic (FM) interfaces, first observed by Meiklejohn and Bean[6] in small particles of Co with oxidized surface. The main feature of this coupling, also known as exchange bias (or exchange anisotropy), is a shift of the center of hysteresis loops from zero field. It has been observed in bilayers with permalloy (FeNi) and an AF material, such as FeMn,[7] CoO,[8] and NiO,[9] and more recently in Fe/FeF_2 bilayers.[10] Roughly speaking, the source of this asymmetry originates from exchange coupling of AF and FM spins at the interface. Cooling the sample in a positive field from above the AF ordering temperature (T_N), the field values required to invert the magnetization of the FM layer (M_{FM}) from $+M_{FM}$ to $-M_{FM}$ and from $-M_{FM}$ to $+M_{FM}$ are different because, besides the Zeeman energy, there exists the exchange coupling between AF and FM spins which depends on the *initial configuration* of AF spins at the interface. Actually, the AF initial configuration produced by the field cooling serves to lock the interactions of individual spins at the interface. The hysteresis loop can be shifted to negative or positive fields, the value of the shift being referred to as the exchange bias field (H_E). Several models have been proposed to explain the sign and intensity of H_E, taking into account different initial AF spin configurations.[12–17] Note that any kind of disorder at the interface such as roughness, alloying, and dislocations tends to decrease the magnitude of H_E.[11]

In the present paper, we report structural and magnetic properties of quartz/Sm(25 nm)/Fe(15 nm) thin films. In the Sm/Fe bilayer it is well understood how the microstructure contributes to the magnetic anisotropy of the film. The Fe layer is composed of small elongated monodomain particles with their major axes parallel. Thus, the magnetic shape anisotropy of individual particles is ''added,'' producing a uniaxial magnetic anisotropy in the film. Such uniaxial anisotropy has been clearly observed from measurements of the coercive field (H_C) at room temperature, varying the angle between the sample easy axis and the applied magnetic field. Furthermore, resulting from exchange coupling the Sm/Fe bilayer exhibits shifted hysteresis loops below 40 K [the (T_N) of Sm is 106 K (Ref. 18)], and the dependence on temperature of H_E was measured. One interesting aspect in this system is the long-range character of the RKKY interaction between the Sm-Sm and Sm-Fe spins near the interface, which has not yet been explored in other AF/FM bilayers.

Films of quartz/Sm/Fe have been grown in an ultrahigh vacuum (UHV) chamber with a base pressure lower than 1×10^{-9} mbar. Both Sm and Fe were evaporated using a commercial Omicrom electron-beam evaporator at a low deposi-

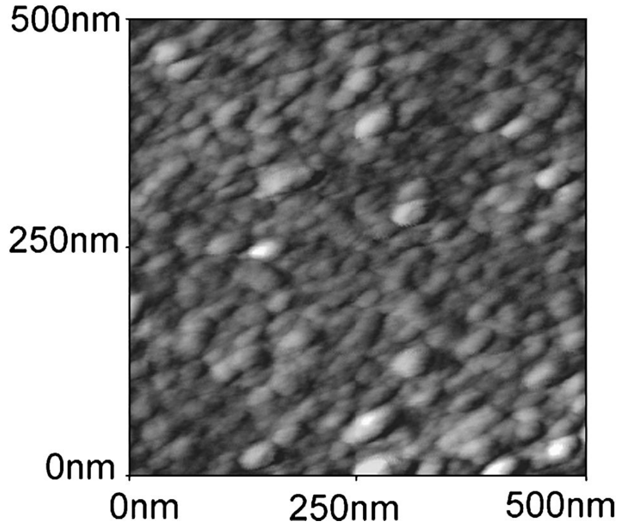

FIG. 1. AFM image of Fe layer. The image shows elongated Fe grains with the mean length and width as 36 ± 12 nm and 21 ± 7 nm, respectively. Note that the major axes are all parallel.

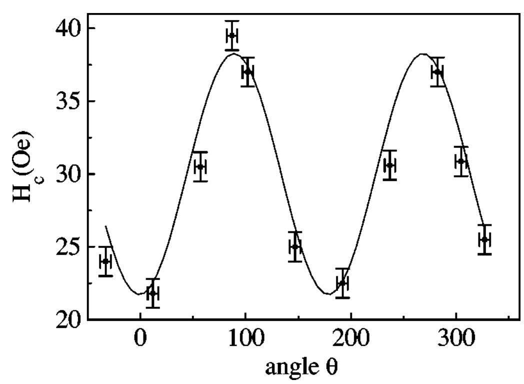

FIG. 2. Measurements of coercive field (H_C) by changing the angle between the magnetic anisotropy axis and the magnetic field applied in the plane of the film. This curve indicates that the magnetic anisotropy is uniaxial. The line is a guide to the eyes.

tion rate (2 ML/min). The quartz substrates were cleaned by immersion in acetone before loading into the chamber, and by annealing at 600 K for 1 h in UHV conditions. First, the Sm layer was evaporated onto the quartz substrate at room temperature followed by a fast heating up to 900 K. Afterwards, the Fe layer was evaporated onto the Sm one at room temperature. In order to induce a uniaxial anisotropy the deposition of Sm and Fe layers was carried out with the vapor beam arriving onto the substrate forming an angle to the normal surface of approximately 5°. In addition, it has recently been shown through the initial growth of Sm on Pt substrates that Sm atoms have great mobility at the surface, favoring the formation of elongated grains.[19] The characterization of films was made *ex situ* through grazing incidence ($\cong 1°$), large angle x-ray diffraction and AFM. The AFM analyses were done using a commercial system (Topometrix, Discoverer model) in the noncontact mode.

Analyzing the (100) and (110) reflections for Sm and Fe layers, respectively, on grazing incidence and rotating the film around an axis perpendicular to the film plane, we observed that the peak intensity exhibits a periodic behavior. This means that the deposition of Sm and Fe layers leads to a preferential growth along a crystallographic orientation forming grains with a textured structure. Atomic force microscopy (AFM) measurements provided the mean size for Sm grains (about 80 nm) and show that the Fe grains are elongated and oriented along a preferential direction (see Fig. 1). The latter grains are 36 ± 12 nm long and 21 ± 7 nm wide with aspect ratio equal to 1.6, approximately. It is interesting to remark that, although the grain size follows a distribution function, the aspect ratio is almost the same for the whole size range.

Magnetization measurements were performed *ex situ* through longitudinal magneto-optic Kerr effect (MOKE) and Supercondicting quantum interference device (SQUID) magnetometry. Our MOKE setup is composed of a diode laser emitting red light (wavelength 670 nm); the polarizer and the analyzer is arranged almost crossed, and the magnetic field is applied in the plane of light incidence. The incident light was linear *p* polarized and lock-in detection with light amplitude modulation was used.

Hysteresis loops were measured using MOKE at room temperature with the magnetic field applied in the film plane and forming an angle (θ) in respect to the direction of minor axes of the Fe grains. We show in Fig. 2 the coercive field (H_C) at room temperature for the field applied in different orientations. H_C follows a sinusoidal dependence on θ, oscillating between 23 and 38 Oe with a periodicity of 180°. In fact, this periodicity agrees with AFM measurements, i.e., the Fe grains exhibit the same uniaxial symmetry. Note that at room temperature the Sm layer is not magnetically ordered, and the magnetic response is attributed to the Fe layer only. Hence, we attributed the observed magnetic anisotropy to the shape anisotropy of the Fe grains. In addition, this idea is supported by the fact that the size of Fe grains observed here is below or of the order of the critical diameter for Fe ($\cong$ 40 nm as mean diameter) below which a magnetic particle becomes monodomain. If the Fe grains are larger than the critical value, magnetic domains will be present inside the grains and no magnetic anisotropy should be observed. In conclusion, the granular textured microstructure is the source of the uniaxial magnetic anisotropy observed in the film.

In the light of the noninteracting monodomain grain model given by the Stoner and Wohlfarth (SW) model[20] one can discuss in more detail the hysteresis loops. The squareness behavior, typical of angles where H_C has the maximum value (see Fig. 3), e.g., $\theta=86°$, is characteristic of magnetization curves of monodomain particles for the magnetic field applied parallel to the easy axis (long axis of the grain). In this case, the magnetization reversal occurs through coherent rotation. On the other hand, for orientations where H_C has the minimum value, e.g., $\theta=190°$, hysteresis loops correspond to the hard axis. In that case, the magnetization rotates up to a critical value, above which the magnetization suddenly turns to the field direction.[21] Note that, according to the SW model, for the magnetic field applied along the hard direction no hysteresis exists. However, in our system, the observed hysteresis may result from the interaction between the grains.

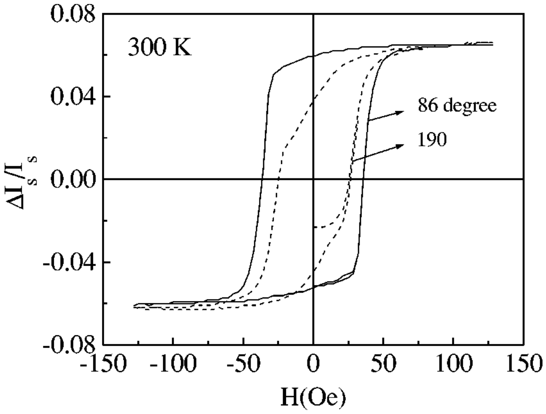

FIG. 3. Hysteresis loop for $\theta=86$ (full line) and 190° (dashed line) measured by MOKE at 300 K.

In Fig. 4 hysteresis loops measured on a SQUID magnetometer (Quantum Design, MPMS XL model) at 5 K for $\theta=86°$ show that the hysteresis loops centers are shifted 96 Oe to negative field (see inset). The same behavior is observed when the sample is cooled with or without magnetic field, at least up to 10 kOe. It probably occurs because upon decreasing the temperature a local field from the Fe layer on Sm atoms at the interface is sufficient to lock in the exchange interaction. In Fig. 4 we show the temperature dependence of H_E plotted in a log-log scale. Note that contrary to other systems exhibiting exchange bias coupling, H_E decreases very quickly, following a power law decay with temperature, $H_E \alpha 1/T^{\gamma}$, where $\gamma=1.03\pm0.08$. Furthermore, the fact that H_E vanishes at the temperature value of 40 K, which is below T_N of Sm (106 K) and not at T_N, as usually occurs in these systems, may be due to either the rapid decreasing of H_E, or to a possible reduction of T_N, caused by the formation of some nonequilibrium alloy at the interface.

Before discussing the origin of the power law temperature dependence of H_E, it is interesting to remark that due to the high reactivy of the rare earths, a source of disorder arises from formation of a Sm-Fe alloy at the interface, which could be enough to destroy a magnetic coupling. From our results, we have not been able to determine the thickness of the region of alloying or disorder in our samples. From our AFM measurements on a Sm surface we have found a mean roughness amplitude of 0.8 nm. However, after Fe deposition an interdiffusion of Sm-Fe ions occurs. Due to the high reactivity of rare-earth metals, one can expect the thickness of alloying to be greater than the roughness of the Sm surface. Nevertheless, the long-range character of the rare-earth spin interaction overcomes the thickness of this disordered region. Thus, it is reasonable to conclude that even with alloying or other sources of disorder at the interface, the Sm-Fe layer coupling interaction still prevails.

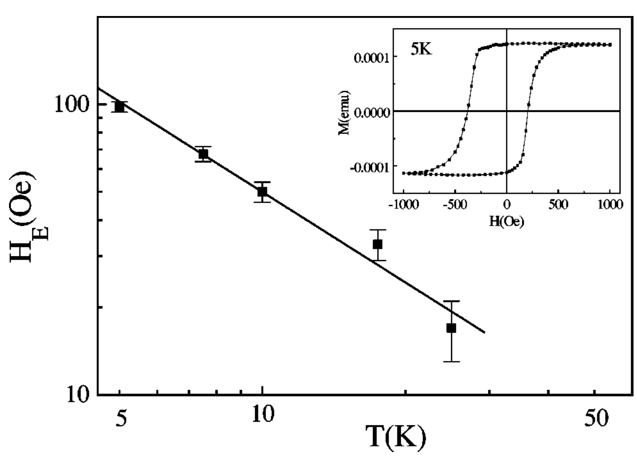

FIG. 4. Exchange bias field (H_E) as a function of temperature in log-log scale. The inset shows a hysteresis loop performed at 5 K, shifted to negative fields.

Cooling the sample from above T_N the majority of Sm spins at the interface will point along the field direction, with some in the opposite direction. In that way the exchange coupling $-J_{Sm,Fe}S_{i,Sm}S_{j,Fe}$ summed for all Sm and Fe spins at the interface is maximum if $S_{i,Sm}$ has the same value for all Sm spins; otherwise the exchange coupling decreases, thus decreasing H_E [$J_{Sm,Fe}$ is the exchange constant between Sm and Fe spins, and $S_{i,Sm(Fe)}$ is the spin variable for a Sm (Fe) spin i]. Note that spatial disorder caused by roughness or alloying also decreases H_E by randomizing $S_{i,Sm(Fe)}$. For a given temperature, if a Sm spin aligns opposite to the field direction, due to long-range Sm-Sm spin interaction other Sm spins will also be antiparallel within a correlation length. Hence, it produces a quick decrease of the exchange coupling with temperature, and therefore of H_E. It is well known that long-range spatial interactions and long-range memory effects exhibit power laws as a signature.[22] *Thus, the observed power-law decrease of H_E with temperature may be interpreted as a consequence of long-range interactions at the interface.*

From the above comments it is natural to consider that the spin configuration at the interface plays a crucial role in the exchange bias. Spin polarized electron diffraction studies have shown the existence of magnetic properties at the surface that differ from the bulk in magnetic thin films.[23] For instance, the magnetic order and the ordering temperature at the surface may be different from those of the bulk, and the same should occur at the interfaces. It has been reported that this effect is more pronounced in rare-earth systems,[24] due to long-range spin interactions. Hence, the knowledge of the spin configuration at the interface is of fundamental importance in the understanding of exchange bias in Sm/Fe bilayers.

Another possible explanation for our results consists of considering the alloy formation at the interface interacting with the Fe layer, forming thus two distinct hard and soft phases as a spring exchange behavior. For instance, recent results have been reported in SmCo alloy/TM bilayers (TM = Fe and Co).[25] In the latter the demagnetization curve exhibits two steps, each corresponding to soft (TM) and hard (SmCo) layer magnetization reversal. In Sm/Fe bilayers, however, such behavior is not observed, but only a shift of the hysteresis loop, as occurs in exchange bias coupling.

In summary, we have shown that in the quartz/Sm(25 nm)/Fe(15 nm) system, Sm and Fe layers grown with a textured granular structure give rise to a uniaxial magnetic anisotropy in the plane of the film. An exchange bias coupling between Sm and Fe layers exists in spite of the Sm-Fe alloying/disorder at the interface, and furthermore, H_E decreases as a power law with temperature. We assume that this is a consequence of the long-range character of Sm-Sm and Sm-Fe interactions at the interface.

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[1] *Ultrathin Magnetic Structures* I, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin,1994), Vol. I, Chap. 5, p. 177.
[2] A. Berger, A.W. Pang, and H. Hopster, Phys. Rev. B **52**, 1078 (1995); M. Gajdzik, T. Trappmann, C. Surgers, and H. von Lohneysen, *ibid.* **57**, 3525 (1998).
[3] L.T. Baczewski, R. Kalinowski and A. Wawro, J. Magn. Magn. Mater. **177**, 1305 (1998); Y. Li, C. Polaczyk, F. Mezei, and D. Riegel, Physica B **234**, 489 (1997); N. Cherief, D. Givord, O. McGrath, Y. Otani, and F. Robaut, J. Magn. Magn. Mater. **126**, 225 (1993).
[4] C. Bryn-Jacobsen, R.A. Cowley, D.F.M. McMorrow, J.P. Goff, R.C.C. Ward, and M.R. Well, Phys. Rev. B **55**, 14 360 (1997), and references therein.
[5] *Ultrathin Magnetic Structures* II, edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin, 1994), Vol. II, Chap. 2, p. 45.
[6] W.P. Meiklejohn and C.P. Bean, Phys. Rev. **102**, 1413 (1956); **105**, 904 (1957).
[7] D.H. Hempstead, S. Krongelb, and D.A. Thompson, IEEE Trans. Magn. **14**, 521 (1978).
[8] T.M. Moran, J.M. Gallego, and I.K. Schuller, J. Appl. Phys. **78**, 1887 (1995).
[9] M.J. Carey and A.E. Berkowitz, Appl. Phys. Lett. **60**, 3060 (1992).
[10] J. Nogues, D. Lederman, T.J. Moran, and I. Schuller, Phys. Rev. Lett. **76**, 4624 (1996).
[11] J. Nogues, D. Lederman, T.J. Moran, Ivan K. Schuller, and K.V. Rao, J. Appl. Phys. **68**, 3186 (1996).
[12] A.P. Malozemoff, Phys. Rev. B **35**, 3679 (1987); **37**, 7673 (1988); J. Appl. Phys. **63**, 3874 (1988).
[13] D. Mauri, H.C. Siegmann, P.S. Bagus, and E. Kay, J. Appl. Phys. **62**, 3047 (1987).
[14] T.M. Moran and I.K. Schuller, J. Appl. Phys. **79**, 5109 (1996).
[15] T. Ambrose, R.L. Sommer, and C.L. Chien, Phys. Rev. B **56**, 83 (1997).
[16] N.C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).
[17] T.M. Hong, Phys. Rev. B **58**, 97 (1998).
[18] W.C. Koehler and R.M. Moon, Phys. Rev. Lett. **29**, 1468 (1972).
[19] H.J. Venvik, C. Berg, A. Borg, and S. Raaen, Phys. Rev. B **53**, 15 964 (1996).
[20] R.C. Stoner and E.P. Wohlfarth, Philos. Trans. R. Soc. London, Ser. A **240A**, 599 (1948).
[21] C. Daboo, R.J. Hicken, E. Gu, M. Gester, S.J. Gray, D.E.P. Eley, E. Ahmad, J.A.C. Bland, R. Poessly, and J.N. Chapman, Phys. Rev. B **51**, 15 964 (1995).
[22] G. Grinstein, in *Scale Invariance, Interfaces and Non-Equilibrium Dynamics*, edited by Mckane *et al.* (Plenum Press, New York, 1995).
[23] H. Hopster, in *Ultrathin Magnetic Structures* I (Ref. 1), Chap. 4, p. 138.
[24] C. Rau, J. Magn. Magn. Mater. **31-34**, 874 (1983); C. Rau, and S. Eichner, Phys. Rev. B **34**, 6437 (1986); C. Rau, C. Jin, and M. Robert, Phys. Lett. A **138**, 334 (1989).
[25] E.E. Fullerton, J.S. Jiang, M. Grimsditch, C.H. Sowers, and S.D. Bader, Phys. Rev. B **58**, 12 193 (1998).