ELSEVIER

Journal of Magnetism and Magnetic Materials 251 (2002) 202–206

www.elsevier.com/locate/jmmm

Thickness-dependent magnetic properties of $Ni_{81}Fe_{19}$, $Co_{90}Fe_{10}$ and $Ni_{65}Fe_{15}Co_{20}$ thin films

S. Ingvarsson[a,*], Gang Xiao[a], S.S.P. Parkin[b], W.J. Gallagher[c]

[a] *Physics Department, Brown University, Providence, RI 02912, USA*
[b] *IBM Research Division, Almaden Research Center, Almaden, CA 95120, USA*
[c] *IBM Research Division, T.J. Watson Research Center, Yorktown Heights, NY 10598, USA*

Received 4 January 2002; received in revised form 17 July 2002

Abstract

We present results of magnetization and magnetic anisotropy measurements in thin magnetic films of the alloys $Ni_{81}Fe_{19}$, $Co_{90}Fe_{10}$ and $Ni_{65}Fe_{15}Co_{20}$ that are commonly used in magnetoelectronic devices. The films were sandwiched between layers of Ta. At room temperature the critical thickness for all the films to become ferromagnetic is in the range 11–13 Å. In $Co_{90}Fe_{10}$ the coercivity and the anisotropy field both depend strongly on layer thickness.
© 2002 Elsevier Science B.V. All rights reserved.

PACS: 75.70.Ak

Keywords: Magnetic thin films; Magnetization; Coercivity; Anisotropy; Magnetoelectronic devices

With the recent interest in magnetoelectronic devices that take advantage of spin-dependent effects, it is important to characterize the magnetic properties of those alloys that are most commonly used in such applications. In particular, for high-density magnetic random access memories (MRAM), it is advantageous to use very thin magnetic films from the standpoint of minimizing the demagnetizing field in the devices and maintaining low switching fields (of a "free layer") required for device operation. In general, having a thin magnetic film does not adversely affect the performance of magnetoelectronic devices. For instance in magnetic tunnel junctions (MTJs), it has been shown that 1–2 monolayers of the ferromagnetic (FM) film at the FM/tunnel-barrier interface suffice to spin polarize the electron tunneling current [1,2]. However, there may be thickness-dependent effects that set limits on the useful thickness range of magnetic films. Properties such as coercivity (H_c) and magnetic anisotropy (which in the case of a uniaxial material can be characterized by, e.g. the uniaxial anisotropy field, H_k) are affected by mechanical strain and crystal symmetry, which are clearly altered at a surface or an interface. When averaged over the entire volume of the film, these properties can therefore be expected to depend strongly on the

*Corresponding author. Present address: IBM Research Division, T.J. Watson Research Center, P.O. Box 218, Room 02-038, Yorktown Heights, NY 10598-0218, USA. Tel.: +1914-945-2292; fax: +1914-945-4440.

E-mail address: sthi@us.ibm.com (S. Ingvarsson).

0304-8853/02/$ - see front matter © 2002 Elsevier Science B.V. All rights reserved.
PII: S 0 3 0 4 - 8 8 5 3 (0 2) 0 0 5 7 7 - 2

volume to surface ratio, i.e. its thickness. Measurements of magnetic moment versus thickness in thin films of ferromagnetic transition metals typically exhibit a linear relationship which is offset from the origin. This is the case for the results displayed in Fig. 1. The offset in Fig. 1 indicates an effective magnetic thickness *smaller* than the actual layer thickness. Primarily two mechanisms have been used to explain such results. The first is that magnetically dead layers at the interface, i.e. the part of the ferromagnetic film nearest to the interface, is nonmagnetic (this was shown to apply to e.g. Ta/NiFe interfaces, see Refs. [3,4]). The second is that the magnetic ordering temperature (Curie temperature, T_c) is lowered in the atomic layers close to the interface (see e.g. Ref. [5]). If the measurement temperature is higher than the T_c of the interface layers this could show as a reduced magnetic moment in experiments where the paramagnetic surface layers are not completely saturated. In the cases above, below a certain film thickness the film either becomes nonmagnetic or paramagnetic, respectively, and useless for magnetoelectronic applications. Therefore, it is extremely important to study the thickness dependence of magnetic properties.

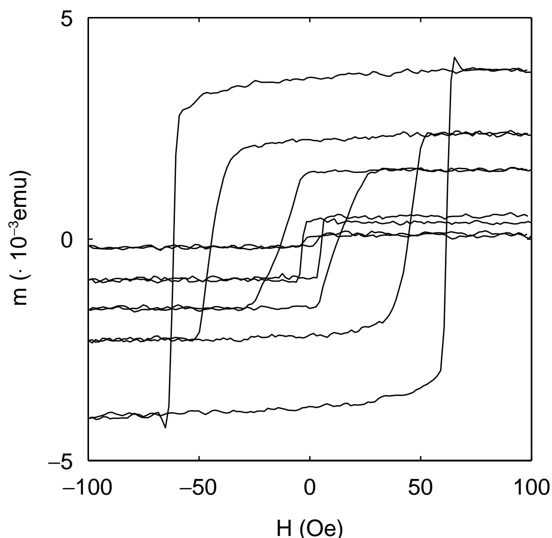

Fig. 1. Magnetic moment as function of applied field for different thickness CoFe films. The thickness values are, in the order of shrinking loops, $t = 60$, 40, 30, 20, 15 Å, respectively. 10 Å thick CoFe did not display hysteresis. The coercivity, H_c, and the anisotropy field, H_k have strong dependence on thickness in these films.

We chose to study three different FM alloys, $Ni_{81}Fe_{19}$, $Co_{90}Fe_{10}$ and $Ni_{65}Fe_{15}Co_{20}$ (to be referred to as NiFe, CoFe and NiFeCo, respectively) that have been used extensively as active magnetic films in giant magnetoresistance (GMR) spin valves and MTJs. Thin films were sputter-deposited on thermally oxidized Si-wafers and have the following layer structure (numbers denote thickness in Å) $Si/SiO_2/50$ Ta/FM/40 Ta, where FM is one of the aforementioned alloys. The FM film thickness ranges between 10 and 100 Å. We used Ta both as a buffer layer and a capping layer. It is known that growing NiFe on a Ta buffer layer results in small grain sizes and strongly (1 1 1) textured layers [6]. The films are grown in an in situ magnetic field of ~ 150 Oe, which determines the direction of the easy-axis anisotropy in the films. The pressure of the Ar sputtering gas was 3 mTorr, and the sputtering rates were within 1–2 Å/s. The vacuum base pressure was 4×10^{-9} Torr before sputtering. Magnetic properties were measured with a vibrating sample magnetometer (VSM).

Fig. 1 shows the magnetic hysteresis loops of one of the three thin film series, the CoFe series, as a function of film thickness. The magnetic field was applied along the easy-axis direction. In all of our samples the hysteresis loops are square like with well-defined switching fields. As shown in Fig. 1, H_c is very sensitive to film thickness in CoFe thin films, increasing with the film thickness. In NiFe and NiFeCo H_c remains essentially unchanged (and small compared to CoFe) as thickness is varied (see Fig. 5). The maximum applied field in these measurements was 100 Oe. In all but the thickest two CoFe films this resulted in a wide saturation region with constant magnetization to within the sensitivity of the apparatus. The magnetization of the two thickest CoFe films leveled off to a constant magnetization above 60 Oe. We estimate the magnetization from an average over 20 points closest to the extreme field values, i.e. in the absolute field range 70–100 Oe. We found that the saturation magnetic moment, m, defined in this way depends linearly on film

thickness, t, according to

$$m = M_s A(t - t_c), \quad (1)$$

where M_s is magnetization, A the film area, and t_c is the critical thickness below which the film is no longer ferromagnetic. Fig. 2 displays the magnetic moment for all three alloys as a function of t. By linear extrapolation to zero moment, we obtained t_c as listed in Table 1. For all these alloys t_c is between 11 and 13 Å. Under the naive assumption (see Ref. [4]) that the two interfaces within each stack of Ta/FM alloy/Ta are equivalent, the nonferromagnetic layer thickness is about 6 Å at each interface.

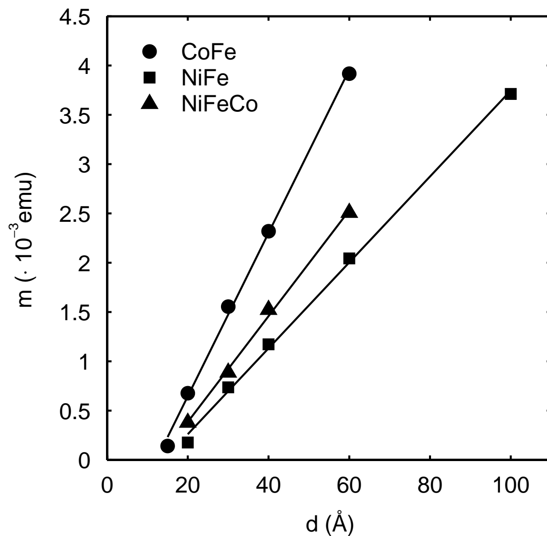

Fig. 2. Magnetic moment as function of layer thickness of our alloys. The roughly linear dependence is indicative of a constant magnetization. The offset from the origin allows determination of the critical thickness for the film to exhibit magnetic long-range order at room temperature.

Table 1
Saturation magnetization, M_s, obtained from the kink field in Fig. 3, and the critical thickness t_c at which our films become nonferromagnetic, i.e. either nonmagnetic [3,4] or paramagnetic [5].

	M_s (emu/cm^3)	t_c (Å)
CoFe	1302	11.2
NiFe	785	12.4
NiFeCo	850	12.8

We estimated independently the magnetization of the thickest magnetic films of each alloy (60 Å CoFe and NiFeCo, respectively, and 100 Å NiFe) by measuring the magnetization perpendicular to the films, the results of which are shown in Fig. 3. The saturation field in such a measurement is equal to the out-of-plane demagnetizing term, $4\pi M_s$, and yields the magnetization independent of film volume or the absolute magnetization calibration of the apparatus. These results are in good agreement with bulk values [7]. We used these results as an exact moment calibration for the data in Fig. 2. Then by correcting the sample volume for nonferromagnetic layers at the interfaces as in Eq. (1), assuming that their contribution to the magnetization is negligible or zero, we obtained the magnetization results shown in Fig. 4. The magnetization of all the three alloys remains constant at least down to a layer thicknesses of 30 Å. The next thickness was 20 Å, at which point there appears to be a slight decrease in the magnetization of the CoFe, and a substantial reduction in the case of NiFe. The 15 Å CoFe film

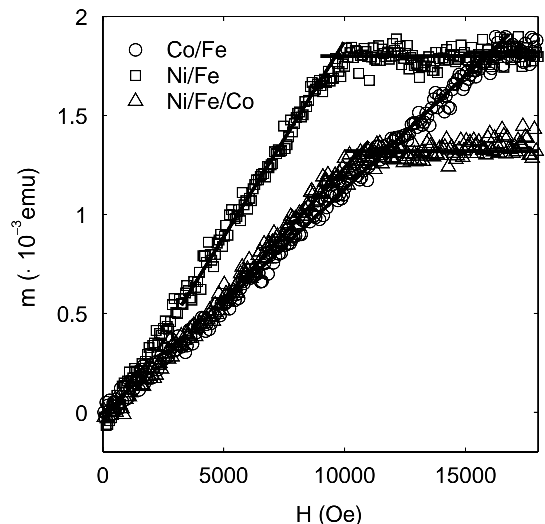

Fig. 3. Magnetic moment perpendicular to the plane of the thickest films of each alloy (60 Å CoFe and NiFeCo, respectively, and 100 Å NiFe). The kink field gives the magnetization, independent of the in-plane geometry of the film, since $H_{kink} = 4\pi M_s$. The lines are a guide to the eye. The magnetization values extracted from these data are shown in Table 1.

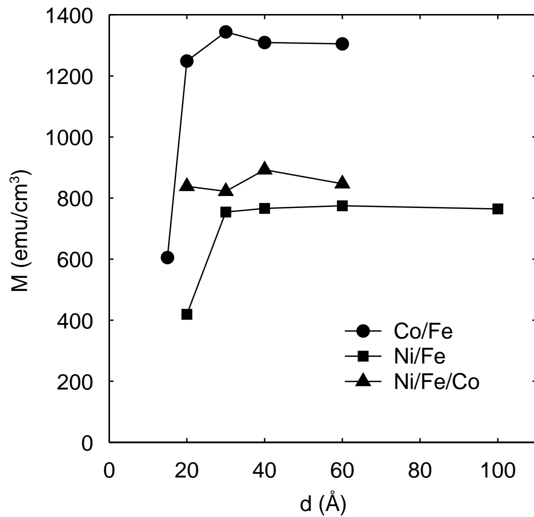

Fig. 4. Magnetization calculated by combining the magnetization determined in Fig. 3 with the data in Fig. 2 and effective magnetic volume.

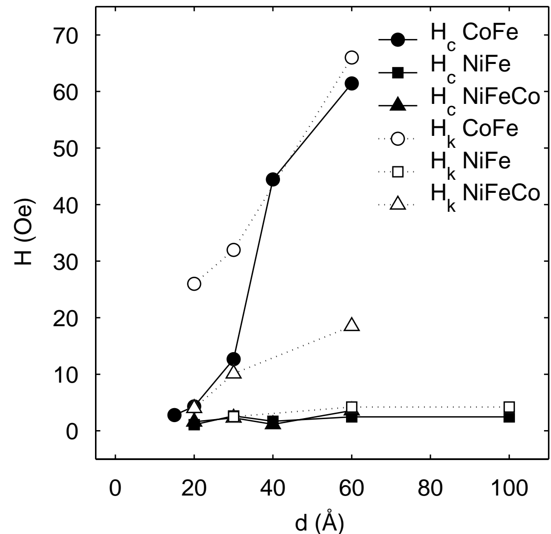

Fig. 5. Coercivity, H_c, and anisotropy field, H_k, as function of film thickness. Both these parameters have a very strong thickness dependence in CoFe. Only H_k has any appreciable dependence on NiFeCo thickness, and neither H_c nor H_k has significant thickness dependence in NiFe.

has a magnetization of less than half of the thick film value. This is of course in contradiction with the initial assumption of a constant magnetization, and equivalent to observing that the points in Fig. 2 do not all fall perfectly on a straight line.

Fig. 5 portrays the thickness dependence of the coercive and anisotropy fields in our samples. The anisotropy field was estimated from magnetization loops taken along the hard axis of the samples, as the field value at which the magnetization saturates. Neither the coercivity nor the anisotropy field of NiFe seem to depend on thickness within our thickness range. The coercivity of the NiFe is in all cases less than 2.6 Oe, consistent with NiFe having a fine grain structure when grown on a Ta buffer layer. The coercive field of NiFeCo is not significantly dependent on thickness, but its anisotropy field however, is a strong function of thickness, reaching 18.5 Oe at a thickness of 60 Å. By far the strongest thickness dependence of these parameters occurs in the CoFe samples. Both the coercive and the anisotropy field become prohibitively large for MRAM applications already at 40 Å (These are large area films, yet to be patterned into small structures. Decreasing their size increases their switching fields). The coercivity of CoFe ranges from 2.8 Oe at 15 Å to 61.5 Oe at 60 Å thickness.

Our results on anisotropy in NiFeCo with Cu buffer and capping layers are in qualitative agreement with those of Wang et al. [8], but their anisotropy field is roughly 3 times that displayed in Fig. 5. Interesting studies have been made on the effects of substrate roughness and of film roughness induced by chemical etching, on the coercivity and anisotropy field of various 3d magnetic alloys [9–12]. These show in general that coercivity increases and anisotropy decreases as roughness is increased. It is difficult to see how roughness of the films could explain our findings. We used identical substrates for all our samples, and deposition conditions remained fixed within each sample series. In our case of ultrathin films, the roughness of the films should not differ much from that of the substrate. Also, in our CoFe samples, H_c and H_k *both* increase as thickness increases, whereas in NiFeCo H_c is more or less constant and H_k increases with thickness. We are led to believe that magnetostrictive effects must be the cause of the increased H_c and H_k observed in CoFe and in H_k in NiFeCo. In bulk CoFe of approximately the

same composition as was used here there is a crossover from positive to negative magnetostriction [7]. If indeed magnetostriction is the cause of the high coercivity this would indicate that the thin film properties depart from the almost zero magnetostriction in the bulk. It is apparent that due to their high coercivity the CoFe films are unsuitable for magnetoelectronic applications requiring low switching fields. NiFe seems to be the best candidate for small magnetoelectronic structures, as the rapidly increasing anisotropy field in NiFeCo may contribute to large switching fields in small structures of that alloy.

The authors would like to thank Yu Lu and David Abraham for helpful discussions. This work was supported in part by DARPA Contract Number MDA-972-99-C-0009.

References

[1] R. Meservey, P.M. Tedrow, Phys. Lett. 238 (1994) 173.
[2] J.S. Moodera, J. Nowak, R.J.M. van de Veerdonk, Phys. Rev. Lett. 80 (1998) 2941.
[3] N. Moghadam, G.M. Stocks, M. Kowalewski, W.H. Butler, J. Appl. Phys. 89 (2001) 6886.
[4] M. Kowalewski, W.H. Butler, N. Moghadam, G.M. Stocks, T.C. Schulthess, K.J. Song, J.R. Thompson, A.S. Arrott, T. Zhu, J. Drewes, et al., J. Appl. Phys. 87 (2000) 5732.
[5] R. Zhang, R.F. Willis, Phys. Rev. Lett. 86 (2001) 2665.
[6] P. Galtier, R. Jerome, T. Valet, in: Polycrystalline Thin Films: Structure, Texture, Properties and Applications, Material Research Society, Pittsburgh, PA, 1994, pp. 417–422.
[7] R.M. Bozorth, Ferromagnetism, IEEE Press, Piscataway, 1978.
[8] D. Wang, J.M. Daughton, K. Bussmann, G.A. Prinz, J. Appl. Phys. 83 (1998) 7034.
[9] H. Jiang, N. Yang, C. Wang, Surf. Sci. 373 (1997) 181.
[10] S. Vilain, J. Ebothe, M. Troyon, J. Magn. Magn. Mater. 157/158 (1996) 274.
[11] V.I. Malyutin, V.E. Osukhovskii, Y.D. Vorobiev, A.G. Shishkov, V.V. Yudin, Phys. Stat. Sol. (A) 65 (1981) 45.
[12] M. Li, Y. Zhao, G. Wang, J. Appl. Phys. 83 (1998) 6287.