Magnetically soft, high moment grain-refined Fe films: application to magnetic tunnel junctions

Georgieva, MT, Telling, ND, Grundy, PJ, Faunce, CA and Jones, GA

http://dx.doi.org/10.1063/1.1776627

Title	Magnetically soft, high moment grain-refined Fe films: application to magnetic tunnel junctions
Authors	Georgieva, MT, Telling, ND, Grundy, PJ, Faunce, CA and Jones, GA
Type	Article
URL	This version is available at: http://usir.salford.ac.uk/368/
Published Date	2004

USIR is a digital collection of the research output of the University of Salford. Where copyright permits, full text material held in the repository is made freely available online and can be read, downloaded and copied for non-commercial private study or research purposes. Please check the manuscript for any further copyright restrictions.

For more information, including our policy and submission procedure, please contact the Repository Team at: usir@salford.ac.uk.

HTML ABSTRACT + LINKS

JOURNAL OF APPLIED PHYSICS VOLUME 96, NUMBER 5 1 SEPTEMBER 2004

Magnetically soft, high moment grain-refined Fe films: Application to magnetic tunnel junctions

M. T. Georgieva, N. D. Telling,[a),b)] P. J. Grundy, C. A. Faunce, and G. A. Jones
Institute for Materials Research, University of Salford, Salford M5 4WT, United Kingdom

(Received 15 October 2003; accepted 6 June 2004)

The effect of N-doping on the microstructure and magnetic properties of thin Fe layers has been employed to construct all Fe-electrode magnetic tunnel junctions that displayed the tunneling magnetoresistance (TMR) effect. Using low nitrogen doses, a reduction in coercivity was achieved due to grain refinement, without a concurrent decrease in the saturation magnetization of the Fe films caused by the formation of crystalline iron nitride phases. It was demonstrated that this N-induced grain refinement can be applied beneficially to control the switching field of the "free" layer in magnetic trilayer structures. In general the ability to control magnetic softness without reducing saturation magnetization will prove important for incorporating high spin-polarized materials into spin valves and TMR devices. © *2004 American Institute of Physics*.
[DOI: 10.1063/1.1776627]

I. INTRODUCTION

In recent years there have been numerous reports on the soft magnetic properties of iron nitride based films, both in single and multiphase form.[1] For many practical applications, such as ultra-high-density data magnetic recording, magnetically soft, high moment pole materials are required. Furthermore, with the development of the giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR) effect in spin valves and magnetic tunnel junctions, respectively, the research interest has been focused on high polarization, magnetically soft materials which can be used as "free" or "soft" magnetic layers in these trilayer structures. The high saturation mangetization of iron makes it suitable for the above applications although its coercivity is not as low as desired owing to its large magnetocrystalline anisotropy constant $K_1 = 4.8 \times 10^5$ erg/cc. The addition of a few percent nitrogen to iron can promote a lower coercivity and magnetostriction without significant reduction of saturation magnetization.[2–5]

In this paper we use the microstructural and magnetic changes observed consequent upon N doping to soften magnetically the free Fe layer in Fe/Al_xO_y/Fe magnetic tunnel junctions (MTJs).

II. EXPERIMENT

In order to establish the correct conditions for doping, single films were first grown on Si(100) wafers by magnetron sputtering in a high vacuum thin-film deposition chamber at a base pressure $\leqslant 2 \times 10^{-8}$ mbar. Undoped, control iron films were deposited in Ar gas with a partial pressure of 2.5×10^{-3} mbar. The nitrogen doped iron layers were obtained with the addition of a low partial pressure of nitrogen to the sputtering gas (the exact conditions for each sample are listed in Table I). The deposition time was fixed, while the nitrogen partial pressure was varied in order to control the amount of nitrogen in the films. The magnetic properties were investigated by vibrating sample magnetometry (VSM) at room temperature. Structural characterization was achieved by a combination of transmission electron microscopy (TEM), electron diffraction and x ray reflectivity (XRR) measurements.

Tunnel junctions were prepared on thermally oxidized Si wafers (SiO_2 thickness 100 nm) with the nominal structure: Si(100)/Fe(300 Å)/{Al(40 Å), O(t s)}/N-doped Fe(600 Å), where t was the oxidation timed varied as 150, 180, 210, and 240 s. The insulating spacer layers of Al_xO_y were prepared by first sputtering a thin (40 Å) Al layer on top of the Fe film and then reacting it with oxygen by direct exposure to an atomic (free radical) oxygen beam from an atom source. The junction area of 10^4 μm^2 was defined using *in situ* changeable contact masks. The oxidized Si wafers prevented current leakage through the substrate. Junction resistance and TMR were measured using a conventional dc four-probe technique.

III. RESULTS AND DISCUSSION

The deposition conditions for a set of six single Fe(N) films are summarized in Table I. Also included in this table I are the film thicknesses, the saturation magnetization M_s (obtained at an applied field of 3 kOe), and the phases identified by the TEM observations. The saturation magnetization values for each sample were obtained by calibrating the VSM with a Ni sample of known dimensions. In order to obtain the correct magnetic sample volume, each film thickness was determined by XRR measurements, while the film area was set by cutting square substrates of fixed 5×5 mm^2 area. Although all samples from this series were deposited for the same time of 5 min, it was found that the thickness of the N-doped iron films decreased slightly with increasing nitrogen content in the films (see Table I). This could be caused by the additional nitrogen gas in the deposition chamber con-

[a)]Present address: Magnetic Spectroscopy Group, CCLRC Daresbury Laboratory, Daresbury, Warrington WA44AD, UK.
[b)]Author to whom correspondence should be addressed; electronic mail: n.d.telling@dl.ac.uk

 © 2004 American Institute of Physics

Downloaded 04 Oct 2006 to 146.87.126.223. Redistribution subject to AIP license or copyright, see http://jap.aip.org/jap/copyright.jsp

TABLE I. Deposition conditions, film thickness, saturation magnetization recorded at $H=3$ kOe, coercivity (for easy axis, e.a., and hard axis, h.a.) and identified crystalline phases for a set of six Fe(N) single films.

Sample name	N_2/Ar gas flow ratio (%)	Film thickness (Å)	M_s(emu/cc)	Coercivity H_c(Oe)	Identified phases
Fe_*c*	0	305±5	1730±30	45	α-Fe
FeN1	1.1	305±5	1865±30	20	α-Fe
FeN2	1.9	290±5	1745±30	14(e.a.), 6(h.a.)	α-Fe
FeN3	3.8	275±5	1640±30	11(e.a.), 6(h.a.)	α-Fe+γ'Fe$_4$N
FeN4	7.7	265±5	1540±30	3	$Fe_{2\text{-}3}N$
FeN5	15.4	245±5	1190±30	2	$Fe_{2\text{-}3}N$

taminating the target surface, resulting in a lower deposition rate and thus a reduced thickness of the N-doped Fe films. The calculated values for M_s using the thicknesses derived from the XRR scans are therefore more accurate than if the M_s values were calculated for all samples assuming the same thickness.

The saturation magnetization measured for the undoped Fe film (sample Fe_*c*, Table I) agrees well with the bulk value (1714 emu/cc). It can also be seen from Table I that the saturation magnetization value M_s showed an apparent increase with an addition of a small amount of nitrogen to the sputtering gas (1.1% N_2/Ar gas flow ratio, sample FeN1), reaching a value of 1865 emu/cc (23.4 kG). Such an increase has been attributed previously to the formation of the $Fe_{16}N_2$ phase.[6,7] However, there was no evidence for the existence of this phase in the TEM diffraction patterns of the samples investigated here (Fig. 1). Explanation of this increase in M_s above that for bulk Fe would require further study. Increasing the nitrogen content in the Ar gas resulted in a continuous decrease of the saturation magnetization for samples FeN3, FeN4, and FeN5, consistent with the presence of the FeN phases identified in these films.

The coercivity was found to decrease monotonically for all N-doped samples as the amount of nitrogen in the Fe films increased (see Table I). The formation of FeN phases in the higher N content (FeN3–FeN5) films, as determined from their electron diffraction patterns (Table I), is likely to be responsible for this decreased coercivity since these have been shown previously to be magnetically soft.[1,8] However, no crystalline phases other than bcc α-Fe were identified from the TEM diffraction patterns for the first two N-doped samples FeN1 and FeN2 (see Table I and Fig. 1). On the other hand, the bright field micrographs (Fig. 1) revealed a significant reduction in the grain size of samples FeN1 and FeN2 (<100 Å) compared to the undoped Fe sample, Fe_*c* (250–350 Å). Therefore the reduced coercivity in the first two N-doped iron films can be associated with this decrease in the grain size. Additionally, sample FeN2 had an anisotropic hysteresis loop, i.e., a weak uniaxial in-plane anisotropy appeared in this sample. Preferential ordering of interstitial nitrogen atoms might be responsible for this weak, induced uniaxial anisotropy as reported previously in FeAl(N) alloy films.[9]

A reduction in coercivity has been observed previously in many FeN based alloys and is believed to be a result of the decrease of the grain size correlated with the increase of the nitrogen content in the film.[5,10–13] In nanocrystalline magnetic films with grain sizes ≤100 Å and ferromagnetic grain boundaries, a low coercivity is often interpreted as the result of averaging the magnetocrystalline anisotropy by the ferromagnetic exchange interaction.[14]

We now consider the inclusion of these magnetically soft (grain-refined) Fe layers in MTJs. As is evident from Table I the N-doped Fe film deposited at 1.9% N_2/Ar gas flow ratio shows the most significant decrease in coercivity without formation of FeN phases and a saturation magnetization value as high as for the pure Fe film. Therefore its spin

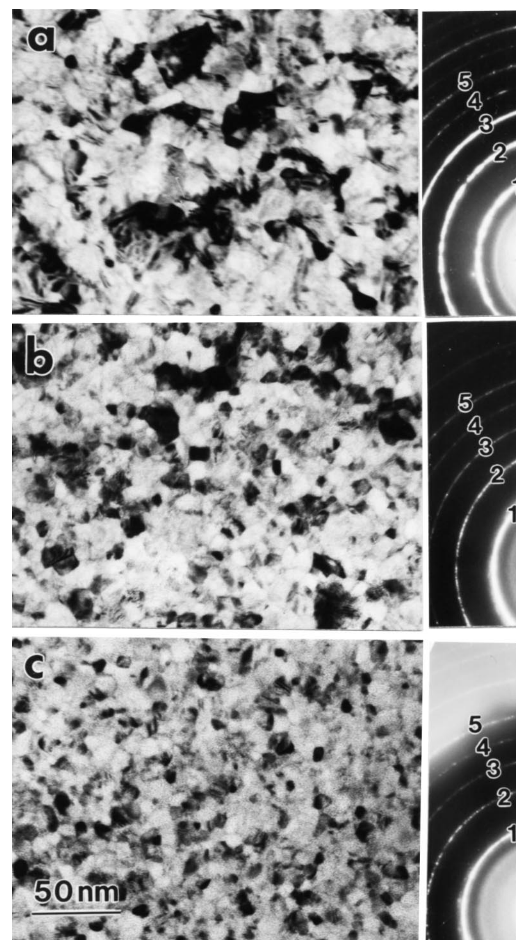

FIG. 1. Bright field micrographs and the corresponding diffraction patterns for (a) undoped Fe film (sample Fe_*c*), and for the N-doped Fe films; (b) sample FeN1, (c) sample FeN2. Insets show the corresponding selected area diffraction patterns. α-Fe rings are designated as reflections from the following planes: 1{110}, 2{200}, 3{211}, 4{220}, 5{310}. The faint innermost rings seen in all samples can be indexed to a thin γ-Fe_2O_3 surface oxide layer.

Downloaded 04 Oct 2006 to 146.87.126.223. Redistribution subject to AIP license or copyright, see http://jap.aip.org/jap/copyright.jsp

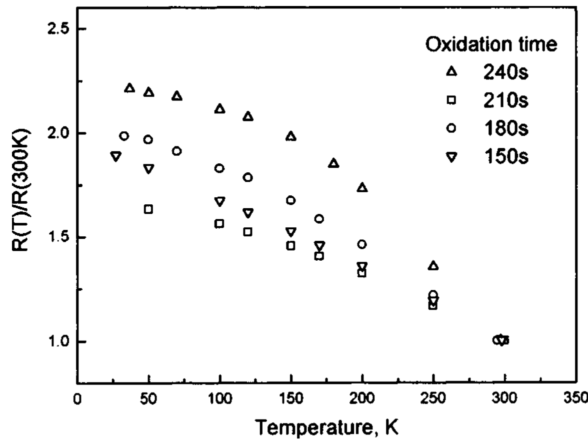

FIG. 2. Normalized junction resistances as a function of temperature for a set of four junctions with the same structure: Fe(400 Å)/Al{40 Å, O(x s)}/N-doped Fe (800 Å) where x is the oxidation time used for the barrier layer.

polarization value is expected to be high and we have chosen this film as the soft (free) magnetic layer in the present junctions.

In our previous work[15] we investigated the magnetization reversal process in trilayer films with the structure: Fe/AlN/Fe (soft). Ferromagnetic interlayer coupling was found in these trilayers which was well explained by the Néel coupling mechanism. By varying the soft Fe layer thickness we demonstrated that the ferromagnetic coupling could be suppressed and a good independent reversal of the magnetic layers obtained in a trilayer system consisting of hard/soft Fe layers. We have applied the same reasoning here to obtain a series of Fe/Al_xO_y/Fe (soft) tunnel junction structures.

All junction samples investigated showed nonlinear current-voltage (I-V) behavior at room temperature with a linear region up to about 100 mV and nonlinear one above it. The I-V curves were well fitted by the Simmons' model[16] allowing the estimation of the barrier parameters (i.e., the barrier width and barrier height). For the range of oxidation times used, the values for the barrier width varied between 14 and 18 Å while those for the barrier height varied between 0.6 and 1.2 eV. Although the values obtained for the barrier height are lower than those typically obtained with plasma oxidation of the barrier,[17–19] they are of the same order as the values reported for the natural and radical oxidation techniques.[20–22]

It has been shown recently[23] that good fits to the measured I-V curves do not preclude the existence of direct conduction via pinholes in the insulating barrier. Tunneling was thus confirmed as the principle transport mechanism by measuring the junction resistance of our samples as they were cooled from 300 to 30 K. All junctions showed a nonmetallic-like temperature dependence (Fig. 2), i.e., the resistance decreased with increasing temperature. The stronger temperature dependence of the junction resistance for all samples as compared to previous measurements[24] can be ascribed to a spin-independent contribution to the junction resistance caused by the possible formation of a nonstoichiometric aluminum oxide (Al_xO_y) insulating layer. This

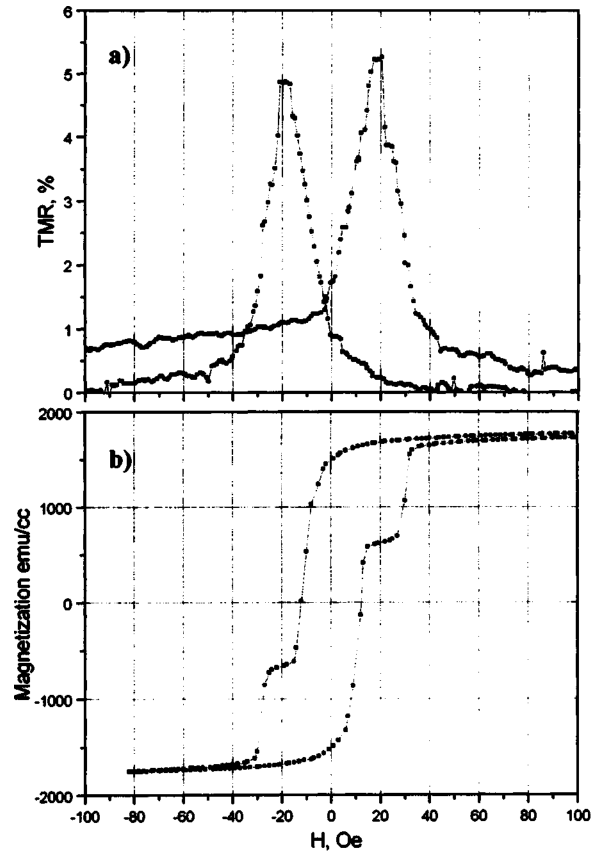

FIG. 3. (a) TMR measured for a junction sample with the structure: Fe(400 Å)/Al{40 Å, O(210 s)}/N-doped Fe(800 Å) and (b) corresponding VSM hysteresis loop of a planar trilayer film deposited simultaneously with the junction.

suggestion is very likely since the deposited Al layer was quite thick and the oxidation process used (atomic free radical oxidation) is a relatively slow and low energy process.

The weakest temperature dependence was found for the junction with the 210 s oxidation time used for the barrier formation, which can be an indication for the optimal oxidation time in this set of samples. Furthermore, TMR measurements performed at room temperature showed the greatest effect for this oxidation time.

The TMR curve for this optimal sample, together with the M-H loop from the simultaneously prepared planar trilayer film with the same structure, are shown in Fig. 3. Although the junction area itself is much smaller than the planar trilayer sample (5×5 mm^2) and the ferromagnetic (FM) electrodes may be thinner due to deposition through the shadow masks (resulting in a stronger Néel-type FM coupling in the junction samples), the planar sample reversal still provides a reasonable illustration of the magnetization reversal process in the junctions. The maxima in the TMR curve [Fig. 3(a)] can be seen to correlate well with the plateau regions in the M-H loop [Fig. 3(b)] which correspond to the antiparallel alignment of the soft (free) and hard FM layers. The lack of a well-defined plateau region in the TMR curve [Fig. 3(a)] can be an indication of ferromagnetic coupling which can lead to a not fully independent switching of the two magnetic layers, and thus a reduction in the measured TMR. However, it can be seen that even when two

Downloaded 04 Oct 2006 to 146.87.126.223. Redistribution subject to AIP license or copyright, see http://jap.aip.org/jap/copyright.jsp

identical FM electrode materials are used, as in this study, grain refinement by N-doping can be used effectively to soften the free layer without adversely affecting its saturation magnetization, thus providing a mechanism for obtaining separate reversal of the FM layers. N-doping could therefore be important for configuring the coercivity of high spin-polarization materials used in spin valves and MTJs.

In this study, although all junction samples showed TMR, the values obtained are lower than expected; between 0.5% and 2% for the underoxidized and overoxidized junctions and between 2% and 5% for those optimally oxidized. In addition to the stronger FM coupling mentioned above, there are two other possible reasons for the low TMR values obtained. First, the method used for the barrier oxidation is a slow and low energy process leading to junctions with much lower resistance than in the case of plasma oxidation. To get a sizeable junction resistance ($>20\ \Omega$) for the given junction area defined by the shadow masks ($10^4\ \mu m^2$), a thick Al layer (~ 40 Å) had to be deposited. It is not possible to oxidize completely and uniformly such a thick layer where the oxygen atoms enter the Al mainly through diffusion (the oxygen atoms have a thermal energy on the order of only 1–2 eV). Therefore the actual barrier layer formed is thinner than the initially deposited Al layer and the rest of the Al is either nonstoichiometric Al_xO_y or pure Al left on the top of the lower Fe layer. In either case this will result in significant reduction of the TMR ratio. The second possibility is that the aluminum oxide formed is not uniform in thickness and only the thinnest regions are properly oxidized and these will be the preferential regions for the tunneling current.

The achievement of low resistance junctions is most desirable with respect to the practical applications of MTJ devices as magnetic random access memory (MRAM) elements or as read heads, where the requirements for the junction-area resistance are less than a few $k\Omega\ \mu m^2$ and less than $10\ \Omega\ \mu m^2$, respectively.[25] The main reason for the low resistance of the present junctions is the use of a large junction area. It is likely that the use of smaller junction areas and thinner, and thus more uniformly oxidized, barriers would result in a significant improvement in the TMR properties of these junctions.

IV. CONCLUSION

It has been shown that N-induced grain refinement that results in a magnetic softening can be used to control the switching field of thin Fe layers incroporated in trilayer structures. It was further demonstrated that the observed microstructural changes caused by the N-doping could be used to control the switching field of the free layer in a magnetic tunnel junction by the fabrication of an all Fe-electrode junction structure that displayed the TMR effect. N-doping could thus prove important for obtaining soft, high spin-polarized materials for applications in magnetoelectronic devices. We further note that N-doping could be used as a method of grain size control in films where microstructure plays an important role in the magnetotransport properties. Examples of these are nanocrystalline magnetic oxide thin films and systems with weak integranular coupling where a reduction in grain size would produce very different changes in magnetic properties to those observed here.

ACKNOWLEDGMENTS

The authors would like to thank the UK EPSRC for support through Grant No. GR/M82776/01 and B. Ashworth for TEM specimen preparation.

[1] J. M. D. Coey, J. Magn. Magn. Mater. **200**, 405 (1999).
[2] N. Terada, Y. Hoshi, M. Naoe, and S. Yamanaka, IEEE Trans. Magn. **20**, 1451 (1984).
[3] C. Chang, J. M. Sivertsen, and J. H. Judy, IEEE Trans. Magn. **23**, 3636 (1987).
[4] S. Wang and M. H. Kryder, J. Appl. Phys. **67**, 5134 (1990).
[5] B. Viala, M. K. Minor, and J. A. Barnard, J. Appl. Phys. **80**, 3941 (1996).
[6] K. H. Jack, Proc. R. Soc. London **A208**, 200 (1951).
[7] M. Takahashi and H. Shoji, J. Magn. Magn. Mater. **208**, 145 (2000).
[8] N. D. Telling, M. J. Bonder, G. A. Jones, C. A. Faunce, and P. J. Grundy, IEEE Trans. Magn. **37**, 2308 (2001).
[9] Yi-K. Liu, V. G. Harris, and M. H. Kryder, IEEE Trans. Magn. **37**, 1779 (2001).
[10] P. C. Kuo, S. S. Chang, C. M. Kuo, and Y. D. Yao, J. Appl. Phys. **83**, 6643 (1998).
[11] S. X. Wang and M. Kryder, J. Appl. Phys. **67**, 5134 (1993).
[12] V. R. Inturi and J. A. Barnard, IEEE Trans. Magn. **31**, 2660 (1995).
[13] G. Qiu, E. Haftek, and J. A. Barnard, J. Appl. Phys. **73**, 6573 (1993).
[14] G. Herzer, IEEE Trans. Magn. **26**, 1397 (1990).
[15] M. T. Georgieva, N. D. Telling, G. A. Jones, P. J. Grundy, T. P. A. Hase, and B. K. Tanner, J. Phys.: Condens. Matter **15**, 617 (2003).
[16] J. S. Simmons, J. Appl. Phys. **34**, 1793 (1963).
[17] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).
[18] S. S. Parkin *et al.*, J. Appl. Phys. **85**, 5828 (1999).
[19] M. Sharma, J. H. Nickel, T. C. Anthony, and S. X. Wang, Appl. Phys. Lett. **77**, 2219 (2000).
[20] T. Mitsuzuka, K. Matsuda, A. Kamijo, and H. Tsuge, J. Appl. Phys. **85**, 5807 (1999).
[21] Z. G. Zhang, P. P. Freitas, A. R. Ramos, N. P. Barradas, and J. C. Soares, Appl. Phys. Lett. **79**, 2219 (2001).
[22] Y. Ando, M. Hayashi, S. Iura, K. Yaoita, C. C. Yu, H. Kubota, and T. Miyazaki, J. Phys. D **35**, 2415 (2002).
[23] B. J. J. Akerman, R. Escudero, C. Leighton, S. Kim, I. K. Schuller, and D. A. Rabson, Appl. Phys. Lett. **77**, 1870 (2000).
[24] C. He Shang, J. Nowak, R. Jansen and J. S. Moodera, Phys. Rev. B **58**, R2917 (1998).
[25] P. P. Freitas, S. Cardoso, R. Sousa, W. Ku, R. Ferreira, V. Chu, and J. P. Condo, IEEE Trans. Magn. **36**, 2796 (2000).

Downloaded 04 Oct 2006 to 146.87.126.223. Redistribution subject to AIP license or copyright, see http://jap.aip.org/jap/copyright.jsp