

Magnetically soft, high-moment, high-resistivity thin films using discontinuous metal/native oxide multilayers

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Multilayers consisting of discontinuous metal layers with native oxide surfaces have been fabricated using $\text{Co}_x\text{Fe}_{100-x}$ alloys ranging in composition from pure Co to pure Fe. For the Fe-containing compositions, the composites are magnetically soft with resistivities in the range of 100 to 2000 $\mu\Omega\text{ cm}$. Mössbauer spectroscopy indicates a magnetically ordered Fe-oxide component, and the magnetic moment of the oxide phase for pure Fe/native oxide multilayers has been determined.

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As data rates in magnetic recording approach gigahertz levels, much effort has been invested in developing magnetically soft, high-moment materials with high resistivity to minimize eddy current effects in magnetic recording heads. With a saturation magnetization in the range of 1400–1700 emu/cc, FeXN (X=Ta, Ti, Cr, etc.) thin films have been the focus of much research.^{1–3} Their magnetically soft properties¹ (coercivity $H_c < 4$ Oe) and uniaxial anisotropy with relatively low anisotropy constant ($H_k < 15$ Oe), combined with a high saturation moment make them of particular interest for recording head applications. However, moderate resistivities⁴ of $\sim 100 \mu\Omega\text{ cm}$ can limit their permeability at high frequencies.⁵

One promising approach to maintain useful magnetic properties while increasing the resistivity has been metal/insulator granular films⁶ consisting of magnetic metallic nanoparticles embedded in a nonmagnetic insulating matrix, such as Al_2O_3 or SiO_2 . However, there is an inherent trade-off in such materials between high resistivity and low coercivity.⁷ For metal volume fractions below percolation, these granular materials are typically superparamagnetic at room temperature. It is only at metal volume fractions approaching percolation that soft properties are obtained, presumably due to exchange coupling amongst nanoparticles over macroscopic regions.⁸ Metallic percolation is, however, necessarily accompanied by decreased resistivity. In addition, as the metal volume fraction must be kept at or below $\sim 50\%$ to achieve high resistivity, the saturation moment of the composite is, at most, about half that of the metal used.

We have developed magnetically soft, high-moment, high-resistivity thin films by fabricating multilayer composites consisting of layers of nanograins of magnetic metal with native oxide surfaces. We have found that this oxide is

magnetic, and its function is threefold. First, it serves to provide an electrical barrier between the grains, thereby increasing resistivity. Second, because of its magnetic nature, the oxide also provides a medium for exchange coupling between the metallic grains, resulting in magnetically soft properties.⁸ Finally, the oxide enhances the moment of the composite beyond what could be achieved in conventional metal/insulator composites.

Layers of $\text{Co}_x\text{Fe}_{100-x}$ with nominal thicknesses varying from 10 to 20 Å were deposited on Si. Previous work has demonstrated that for thicknesses in this range, the layers consisted of isolated nanoparticles for the thinner layers and chains of particles for the thicker layers.⁹ Films were dc sputtered in 2 mTorr of Ar using alloy targets. The depositions were performed in an external dc magnetic field of ~ 140 Oe, which defined an easy axis. Each layer was exposed for 10 s *in situ* to an oxygen flow of 2.0 sccm, yielding an oxygen partial pressure of $\sim 7 \times 10^{-5}$ Torr. The oxygen flow was then terminated, and followed by a pause for 60 s before depositing the next layer, which allowed the oxygen pressure to drop to its background level. This was repeated until the desired film thickness was reached. A final 50 Å SiO_2 capping layer was deposited to prevent further oxidation. The notation $[\text{Co}_x\text{Fe}_{100-x}(t)/\text{oxide}]_N$ denotes a structure in which each of the N unit layers consists of a nominal thickness t of $\text{Co}_x\text{Fe}_{100-x}$ which has been oxidized using the prior procedure.

Samples were characterized structurally by cross-sectional transmission electron microscopy and x-ray diffraction. Magnetic properties were studied using an alternating gradient magnetometer, a superconducting quantum interference device magnetometer, and conversion electron Mössbauer spectroscopy (CEMS). Resistance measurements were performed using a conventional four-point technique.

Figure 1 shows a cross-sectional electron micrograph of $[\text{Co}_{50}\text{Fe}_{50}(20\text{ Å})/\text{oxide}]_{10}$. The regions of light and dark

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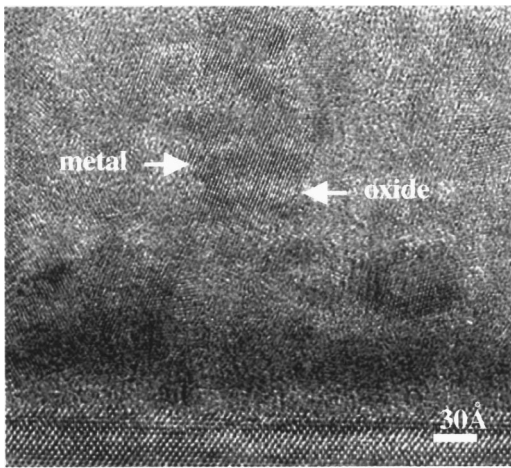


FIG. 1. Cross-sectional electron micrograph of $[\text{Co}_{50}\text{Fe}_{50}(20 \text{ \AA})/\text{oxide}]_{10}$. The dark regions are the metallic layers, and the light regions are the oxide. Note the lattice fringes passing through several layers.

contrast correspond to the oxide and metal, respectively. The roughness of the metal/oxide interfaces confirms the granular nature of the metal layers. Lattice fringes are observed passing through several layers indicating epitaxial growth at some metal/oxide interfaces.

Throughout the alloy composition range, hysteresis loops with well-defined easy and hard axes with low hard axis coercivities are obtained. As an example, Fig. 2 shows easy and hard axis loops for $[\text{Co}_{50}\text{Fe}_{50}(20 \text{ \AA})/\text{oxide}]_{10}$. The easy axis lies along the deposition field direction, and the easy axis loop is very square with $H_c = 11 \text{ Oe}$. The remarkably low hysteresis in the hard axis loop would be especially important for head-shielding applications.

Figure 3(a) compares the coercivity (H_c) of the multilayer films to continuous metal films of the same total nominal thickness. For pure Co, the multilayer structure has a higher coercivity than the continuous 200 Å film. However, near 10 at.% Fe a crossover occurs, and the oxidized multilayer structure has a substantially reduced coercivity compared to the continuous film. This result suggests that the magnetic properties of the oxide and its coupling to the metal are important in determining the coercivity of the composite.

The room-temperature saturation moment of the multilayer films, though decreased from the bulk value,¹⁰ remains high, as seen in Fig. 3(b). The total film thickness

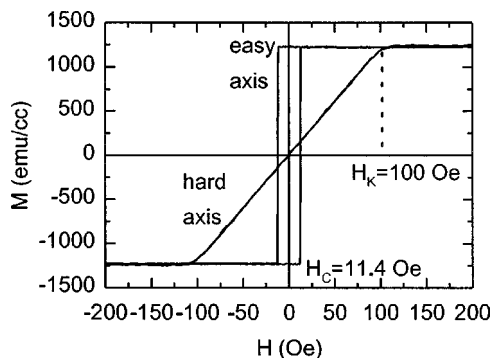


FIG. 2. Easy and hard axis hysteresis loops for $[\text{Co}_{50}\text{Fe}_{50}(20 \text{ \AA})/\text{oxide}]_{10}$. Note the squareness of the easy axis loop, and the low hard axis coercivity.

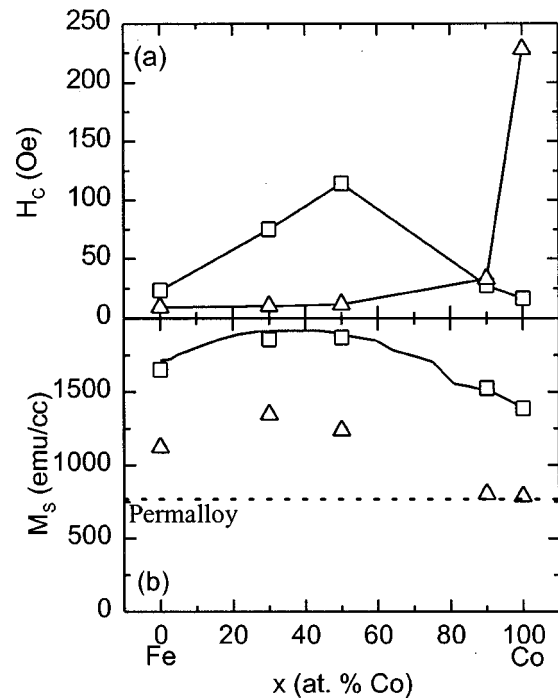


FIG. 3. (a) Coercivity and (b) room temperature saturation moment of $[\text{Co}_x\text{Fe}_{100-x}(20 \text{ \AA})/\text{oxide}]_{10}$ (Δ), compared to continuous 200 Å films (\square). In (b), the bulk saturation moment of $\text{Co}_x\text{Fe}_{100-x}$, taken from Ref. 10, is indicated by the solid curve. The saturation moment of Permalloy is included for reference.

(which is greater than the thickness of the metal deposited due to incorporation of oxygen) was measured using low-angle x-ray diffraction, and used in calculating saturation moments. The saturation value of 1350 emu/cc for $[\text{Co}_{30}\text{Fe}_{70}(20 \text{ \AA})/\text{oxide}]_{10}$, which is the highest moment of the samples studied, is substantially greater than the value of $\sim 800 \text{ emu/cc}$ for permalloy.

The origin of the in-plane anisotropy visible in Fig. 2 is currently under investigation. The anisotropy field H_k of the $[\text{Co}_{50}\text{Fe}_{50}(t)/\text{oxide}]_N$ films is independent of t in the range 10–20 Å. In addition, for $t = 20 \text{ \AA}$, H_k varies linearly with metal composition, ranging from 10 Oe for Fe-based multilayers, to 240 Oe for Co-based multilayers.

All $[\text{Co}_x\text{Fe}_{100-x}(20 \text{ \AA})/\text{oxide}]_{10}$ films show a room-temperature resistivity of $\sim 120 \mu\Omega \text{ cm}$. The resistivity for films with this layer thickness is metallic, as inferred from the temperature dependence. However, as t is decreased and individual layers become more discontinuous, the resistivity increases, as shown in Fig. 4. For $t < 16 \text{ \AA}$, the conductivity becomes thermally activated. For the samples with the thinnest layers, the room-temperature resistivity of $\text{Co}_{50}\text{Fe}_{50}$ -based films exceeds $600 \mu\Omega \text{ cm}$, which is comparable to values obtained in conventional metal/insulator composites with soft magnetic properties.^{7,11,12} The Fe-based multilayers have even larger resistivities.

CEMS was used to investigate the magnetic and chemical environments of the Fe. A detailed presentation of this work will be published elsewhere, but some of the results are noted here. For all Fe-containing multilayers, the CEMS spectra contained magnetically split Fe^0 (metallic) and Fe -oxide peaks, along with a small amount of nonmagnetic Fe -oxide, with Fe valences determined from the isomer shifts. Because this measurement is only sensitive to Fe, we will

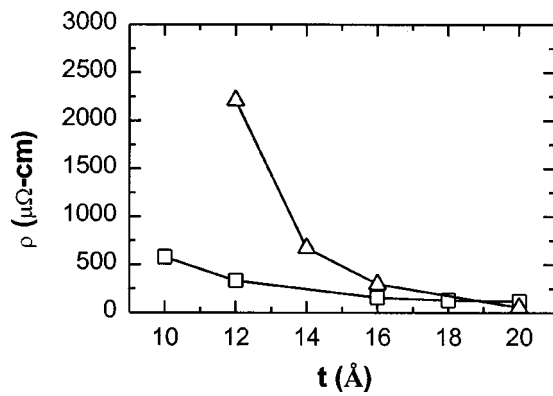


FIG. 4. Room-temperature resistivity of $[\text{Co}_{50}\text{Fe}_{50}(t)/\text{oxide}]_N$ (□) and $[\text{Fe}(t)/\text{oxide}]_N$ (Δ) where t is varied while N is adjusted to keep the total nominal metal thickness at ~ 200 Å.

focus on the $[\text{Fe}(t)/\text{oxide}]_N$ multilayers. CEMS spectra were fitted with metal and oxide subspectra permitting accurate determination of the metallic iron fraction. For the $[\text{Fe}(t)/\text{oxide}]_N$ multilayers, the metallic iron subspectrum has essentially the hyperfine field of bulk α -Fe, and is therefore assumed to have the moment of bulk iron. By using the fraction of Fe as Fe^0 from CEMS and the measured saturation moment, the moment contributed by the oxide phase (or combined phases) can be found. The fraction of Fe as Fe^0 , composite moment as a fraction of bulk α -Fe moment, and calculated moment of the oxide phase are tabulated in Table I for films of several Fe layer thicknesses. As the total amount of iron is known beforehand, the units of moment are $\text{emu}/(\text{cc of Fe})$. In these units, magnetite (Fe_3O_4) has a moment of $1020 \text{ emu}/(\text{cc of Fe})$, and can be used as a reference. It is clear that for all the layer thicknesses, the moment of the composite exceeds that expected from the fraction of Fe^0 .

TABLE I. Fraction of Fe as Fe^0 , moment as a fraction of bulk α -Fe, and calculated moment of oxide phases for $[\text{Fe}(20 \text{ Å})/\text{oxide}]_N$ with $t = 10, 14$, and 20 Å.

t (Å)	Fe^0 fraction	M_s/M_s^{bulk}	M_s^{oxide} (emu/cc of Fe)
20	0.55	0.80	963
14	0.37	0.65	755
10	0.25	0.35	178

This excess moment is attributed to the magnetically split oxide component observed in the CEMS spectra.

Since the native oxide formed in this manner is magnetic, exchange coupling between the metal and the oxide is expected. Thus, these multilayers consist of magnetic metallic nanograins exchange-coupled through magnetic native oxides. Hence, the soft magnetic properties of the discontinuous metal/native oxide multilayer structure can be explained in terms of the random anisotropy model first applied to nanocrystalline materials by Herzer.⁸ This model is valid when the exchange coupling between grains with randomly oriented anisotropy axes is strong enough that the magnetic exchange length L_{ex} is larger than the typical grain size. In this case, the effective anisotropy K_{eff} of the material is the local anisotropy K reduced by the factor \sqrt{n} , where n is the number of grains in the exchange-coupled volume L_{ex}^3 . This lower “average” anisotropy leads to a decreased coercivity.

In summary, we have demonstrated the feasibility of magnetically soft, high-moment, high-resistivity thin films using a composite metal/native oxide structure. The native oxide formed in this way is magnetically ordered, and for pure Fe-based films, has a moment approaching that of magnetite for samples with thicker metal layers. When the iron content of the alloy is greater than $\sim 10\%$, the composite has a much lower coercivity than bulk metal films, a well-defined easy axis, and a remarkably reversible hard axis loop. Resistivities exceeding $2000 \mu\Omega \text{ cm}$ are achieved for the iron-rich films.

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¹ Y.-K. Liu and M. H. Kryder, Appl. Phys. Lett. **77**, 426 (2000).

² B. Viala, M. K. Minor, and J. A. Barnard, J. Appl. Phys. **80**, 3941 (1996).

³ L. Varga, H. Jiang, T. J. Klemmer, W. D. Doyle, and E. A. Payzant, J. Appl. Phys. **83**, 5955 (1998).

⁴ Y. Chen, S. Hossain, C. Qian, M. Miller, and H.-C. Tong, J. Appl. Phys. **85**, 4562 (1999).

⁵ V. R. Inturi and J. A. Barnard, J. Appl. Phys. **81**, 4504 (1997).

⁶ H. Fujimori, S. Mitani, S. Ohnuma, T. Ikeda, T. Shima, and T. Masumoto, Mater. Sci. Eng., A **181/182**, 897 (1994).

⁷ M. Ohnuma, K. Hono, H. Onodera, S. Ohnuma, H. Fujimori, and J. S. Pedersen, J. Appl. Phys. **87**, 817 (2000).

⁸ G. Herzer, IEEE Trans. Magn. **26**, 1397 (1990).

⁹ S. Sankar, Ph.D. thesis, University of California, San Diego, 1999.

¹⁰ P. Weiss and R. Forrer, Ann. Phys. (Paris) **10**, 357 (1929).

¹¹ T. Morikawa, M. Suzuki, and Y. Taga, J. Magn. Soc. Jpn. **23**, 234 (1999).

¹² J. Huijbregtse, F. Roozeboom, J. Sietsma, J. Donkers, T. Kuiper, and E. van de Riet, J. Appl. Phys. **83**, 1569 (1998).