Correlation between domain behavior and magnetic properties of materials

by

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1. INTRODUCTION AND BACKGROUND

Correlation between length scales in the field of magnetism has long been a topic of intensive study. The long-term desire is simple: to determine one theory that completely describes the magnetic behavior of matter from an individual atomic particle all the way up to large masses of material. One key piece to this puzzle is connecting the behavior of a material's domains on the nanometer scale with the magnetic properties of an entire large sample or device on the centimeter scale.

The task of explaining the bulk properties of both ferromagnetic and paramagnetic materials was, in fact, the driving force for the initial formulation of domain theory. The initial debate was over the existence of atomic magnetic moments themselves, with evidence eventually accumulating in favor of what were then called "molecular magnets" [1]. In fact, Ampere postulated that these molecular magnets were due to atomic electrical currents similar to electromagnets, about 75 years before the discovery of the electron [1] and that these currents were permanent even without an external field applied. Over time, pieces to the puzzle began falling into place, including an explanation of paramagnetism based on atomic moment theory by Langevin [2], discovery of the discontinuous nature of the hysteresis loop by Barkhausen, formulation of the idea that a "magnetic" material might have saturated domains oriented randomly to add to zero when demagnetized, and finally in the 1920's and 1930's the idea of domains with finite transition regions called walls, consisting of magnetic moments whose orientations varied across the length of the wall [3].

The actual magnetic domain structures are due to the need for the magnetic material's desire to minimize magnetostatic energy at the cost of formation of the domain walls. As a

Fig. 1: Rotation of individual moments within a 180° domain wall [1].

ferromagnetic material consisting of one bulk domain will have the energy

$$E = \frac{\mu_0}{2} N_d M^2 \quad (\text{where } N_d \text{ is the}$$

demagnetizing factor and M the magnetization), the natural response of the system will be to minimize M [1]. This is accomplished via large groups of collinear moments, usually pointing in favorable

crystallographic directions, with domain walls in which the magnetization vectors rotate from one group's direction to another. If the material is demagnetized or has spontaneously magnetized passing through a Curie point, the total magnetostatic energy of the system can be zero, at the cost of the energy bound in the force required to maintain misorientation of moments in the domain walls. Moment rotation usually increments toward vectors 180° or 90° from the original, as seen in Fig. 1, with the width and number of walls dependent on the strength of exchange and strength of crystalline anisotropy [1]. As many of the interesting properties of ferromagnetic materials derive from the movement, creation, or annihilation of these domain walls, there should be a strong correlation between what is seen at the domain scale and bulk magnetic measurements such as hysteresis curves.

Unfortunately, the initial difficulty of domain imaging and the inherently complicated nature of domains have both retarded progress in their understanding. Initial attempts at viewing domains involved dispersion of ultra-fine iron or magnetite particles on a polished surface, a process that soon evolved into the distribution of a magnetic particle colloid instead of powders. The first published work using this idea came from Bitter [4] and has since been known as the Bitter method. The resolution of the technique has been as good or better than the optical microscopes used to view the patterns, but neither the sensitivity nor repeatability of the technique are reliable and often depend on the skill of the individual researcher. Also, attempts to quantify results by counting particles at boundaries and using complicated models largely failed [5]. A more practical method of imaging domains takes advantage of the Kerr effect, where polarized light waves will have their direction of polarization rotated by magnetic fields at the surface of magnetic materials. In Kerr microscopy, the direction and magnitude of rotation will depend on the direction of the magnetic field with respect to the direction of the incident light beam. This fact that has led to the development of one of the first quantitative methods for domain analysis by Hubert and co-workers [5]. This method

Fig. 2: a) A Kerr micrograph digitally enhanced in b) and analyzed for domain direction in c) [5].

involves taking two misoriented images and digitally processing them together to find both

direction and magnitude of the domains seen. Digital techniques for image processing have also made it simpler to remove artifacts from surface imperfections and enhance contrast in micrographs that are often based on rotations

of less than twenty minutes of arc. An

example of a Kerr micrograph and

subsequent digital enhancements are shown

in Fig. 2. Kerr microscopy has advantages of

large range of possible magnifications,

speedy imaging, and, soon, quantification,

but sample preparation time is long,

equipment complicated and difficult, and resolution relatively limited. Other imaging

techniques include modifications on or use of electron scattering, transmission electron

microscopy (Lorentz microscopy), the Faraday effect on transmitted polarized light, X-rays,

and neutron scattering [5], but all have severe limitations or require extensive sample

preparation. An example of Lorentz microscopy, often used due to the unmatched resolution

of TEM, is shown in Fig. 3.

One of the more useful new tools in the

study of magnetic domain structure has

been the magnetic force microscope, or

MFM. Generally, an MFM involves a

magnetically coated stylus that is

Fig. 3: A Lorentz micrograph permalloy showing "ripple" texture perpendicular to the average domain magnetization [5].

Fig. 4: Photo of an AFM/MFM head and assembly

dragged or tapped across the surface of a solid sample, with the surface stray field gradient causing a force on the tip and providing the contrast. The first such device was designed by Martin and Wickramasinghe at IBM and rastered a magnetic filament across an area of a sample to generate an image, with a resolution of about 100 nm [6]. Current devices, such as the Dimension 3100 AFM/MFM shown in Fig. 4, obtain topographical information by scanning the surface in a fashion similar to atomic force microscopy, then raising the tip a set

Fig. 5: Sketch representation of creating a magnetic force microscopy image.

height above the surface and rescanning. Thus the topology of the sample can be subtracted from the magnetic image and relatively rough sample surfaces can be tolerated [7]. A sketch representation of this process is shown in Fig. 5. Included

are the basic components of the device, including a laser and split photodetector to determine the z -deflection of the tip and, correspondingly, the surface. It should be noted that this strategy does not allow imaging of domains with field gradients oriented in the horizontal sample plane, and that the force on a magnetically coated tip providing contrast is from the vertical component of the stray field gradient (∇H). This makes domain imaging of thin films, where the preponderance of domains are often in plane due to shape anisotropy, more difficult, and in general analysis of any domain imagery slightly more complicated.

However, the end result is much the same as that of scanning electron microscopy topographical imaging (where the contrast is also dependent on a gradient) without the disadvantage of a spatially limited detector. Unfortunately, one problem with using ultra-

sharp stylus tips coated with thin films is the irreproducibility of film thickness and accompanying irreproducibility of contrast from tip to tip. Thus, as long as quantitative measurements of the contrast are not desired, images obtained using this technique will both spatially and qualitatively match the physical picture of the sample domains and can be used by the researchers accordingly to interpret results of measurements in terms of domain structures.

The imaging possibilities of this new instrument were quickly realized. The spatial resolution easily reached the tens of nanometers scale even with rough samples. Some quite striking images have been taken over the last few years, including those shown here in Figures 6-8 [7]. As can easily be seen, many domain features that would have required complicated electron or Kerr microscopy techniques to image can be obtained in little time and with less difficult analysis. In fact, the open nature of the stage makes it possible to add new capabilities to the instrument, such as applied field, temperature control, or other needed capabilities.

Fig. 6: MFM image of a terfenol fracture surface. 75 μm scan by D. G. Lord. [7]

Fig. 7: Magnetic bubbles and stripes in an 8 μm thick garnet film. 100 μm scan by R. M. Westervelt.[7]

Fig. 8: MFM image of domains spreading from small regions of a Co/Pt multilayer that have low anisotropy compared to the rest of the film. 10 μm scan by L. Folks. [7]

2. CASE STUDIES: INTRODUCTIONS AND EXPERIMENTAL METHODS

Due to the varied nature of domain structures, as demonstrated in the previous section, it was not expected that one experiment or set of similar samples would provide enough information to draw broad conclusions. Even among the same material, geometry, coercivity, exchange, anisotropy, temperature, and geometry can significantly affect domain observations. Five different case studies, therefore, have been investigated, discussed, and compared in an attempt to discover trends and consistent patterns, with the end goal of clarifying relationships between domain structures and other magnetic measurements.

2.1 CASE STUDY 1: Stress in an FeSiAl thin film resulting in stripe domains

As-deposited FeSiAl films sputtered in Ar usually have high coercivity, low permeability and large out-of-plane magnetic anisotropy [8]. However, in hard disk drive read heads where they are used for shielding (see Fig. 9), their desired properties are exactly the opposite and are usually achieved by a post-deposition anneal [9, 10]. The drawback to this anneal is its relatively high temperature of $\sim 450^{\circ}\text{C}$, which is well above the temperature suitable for annealing of the entire read head assembly. The necessity to separate the process results in a degradation of the overall performance of the device. A deposition process providing desired soft magnetic properties in FeSiAl without annealing is therefore desirable, and investigations to this purpose discovered that nitrogen additions to the sputtering gas significantly affected film microstructure and magnetic properties in a way that can be useful for this application [11].

Fig. 9: Diagram of magnetic data, with a read/write head and parallel recording medium.

Several FeSiAl(N) films of varying N_2 partial pressure were deposited using radio frequency diode sputtering. An alloy target of Sendust composition (85 wt.% Fe, 10% Si, and 5% Al) was used to deposit films on (100) Si wafers with 300 nm thermally grown SiO_2 . A constant forward power of 1.43 W/cm^2 , fixed deposition time, and fixed combined pressure of Ar and N_2 gases were used to produce $1.7 \mu\text{m}$ films. The partial pressure of N_2 was set to values of 0, 1%, 2%, 3%, 4%, 5%, and 10%. Characterization of the films included vibrating sample magnetometry (VSM), TEM and SEM microstructural and compositional analysis, stress analysis, and magnetic force microscopy. Stress analysis was accomplished by measuring the curvature of long strips on an atomic force microscope, using the compensated z position of the tip at contact and subtracting from the center to determine deflection. Equation (1) describes the stress in terms of the deflection, lateral position, substrate and film thicknesses, film and substrate moduli, and Poisson's ratio of the substrate, and Fig. 10 illustrates the bowing of the substrate due to the film deposition.

$$\sigma = \frac{yE_s D^2}{3x^2 t(1 - \rho_s)} \left(1 - \frac{E_f t}{E_s D} \right) \quad (1)$$

In equation (1), y is the deflection, x is the lateral position, D is the substrate thickness, t is the film thickness, E_s is the Young's modulus of the substrate, E_f is the Young's modulus of the film, and ρ_s is the Poisson's ratio of the substrate.

Fig. 10: Diagram of substrate bowing under stress from film deposition.

For the *in-situ* applied field measurements, an electromagnet capable of producing an in-plane field up to about 56 kA/m (700 Oe) was mounted on the sample stage.

Demagnetization of the samples was accomplished by applying an ac field with decaying amplitude along the desired applied field direction.

2.2 CASE STUDY 2: Magnetization reversal in CoFeHfO films

CoFeHfO films have received considerable attention because of their combination of soft magnetic properties and high-frequency characteristics. It has been reported that CoFeHfO films deposited by reactive sputtering under a dc magnetic field typically have an M_s value exceeding 1T, a coercivity of a few hundred A/m and a high electrical resistivity of the order of magnitude of $600\mu\Omega\text{m}$ [12]. These films were found to contain Fe (or Co-Fe) rich bcc

nanograins and an amorphous matrix containing a large amount of Hf and O [12]. The matrix provides a low conductivity barrier to the long range conduction of current, allowing for very high frequency performance due to suppressed eddy current loss, while the nanograins allow for a relatively high permeability of around 150 along the hard axis. The low loss factor of these films remains constant up to hundreds of megahertz, making CoFeHfO films a promising candidate for high frequency device applications.

A CoFeHfO film (800 nm thick) and a CoFeHfO (10 nm) film overcoated with CrSi (10 nm) were used in this study. The CoFeHfO layers of the samples were deposited by reactive rf-sputtering using an Ar + O₂ atmosphere onto Si (100) substrates with 200 nm of surface SiN. These films were annealed at 250°C for one hour in a magnetic field to induce an in-plane uniaxial anisotropy. Magnetization curves were measured from 7mm by 7mm samples using VSM along both the easy and hard axes of magnetization. Studies of the domain structure and magnetization reversals were made using MFM with tips magnetized perpendicular to the sample plane and the electromagnetic stage described above. Two series of MFM images were taken from each sample under various fields (up to 43.8 kA/m) applied *in-situ* along the easy and hard axes of magnetization of the films.

2.3 CASE STUDY 3: Anisotropy in a Complex Magnetic Material

Gd₅(Si_xGe_{1-x})₄ has recently received much interest due to its extraordinary response in several magnetic and electronic properties during changes in temperature and magnetic field. These include colossal magnetostriction, giant magnetoresistance, and giant magnetocaloric effect. Gd₅(Si_xGe_{1-x})₄ undergoes a magnetic-crystallographic transformation at a Curie

temperature which varies from ~40 K to 320 K dependent on the Si to Ge ratio. During the transformation the material exhibits changes in strain as high as 10^4 parts per million, magnetoresistance of about 25%, and the largest magnetocaloric effect (an adiabatic temperature change when magnetized) to date

Fig. 11: Crystal structure of $\text{Gd}_5(\text{Si}_2\text{Ge}_{1-x})_4$ in both low temperature and high temperature phases. Blue atoms are Gd atoms and red and yellow are Si and Ge, which are often interchangeable.

[13]. The phase transition is a magnetic-martensitic transformation from a paramagnetic-monoclinic crystal structure at higher temperatures to a ferromagnetic-orthorhombic crystal structure at lower temperatures and involves shear of sub-nanometer atomic layers in a complex crystal lattice through reversible breaking and reforming of covalent Si(Ge)—Si(Ge) bonds between the layers [13]. A diagram of the crystal structure can be seen in Fig. 11.

Since initially the magnetic structure of this material was unknown, the assumption had been that the magnetic moments on the Gd atoms in the ferromagnetic state were nearly perpendicular to the b-axis, akin to magnetic structures observed in related Tb_5Si_4 and Tb_5Ge_4 compounds. However, exploratory single crystal MFM measurements indicated

differently, so more comprehensive characterization was initiated, including temperature dependent VSM and MFM.

VSM measurements were collected using a $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ single crystal cube with all three axes identified using x-ray diffractometry. The sample temperature was maintained below the Curie point/transition temperature using a dry ice/ethanol mixture and hysteresis loops measured along the three principal axes of the crystal using a magnetizing field of maximum amplitude of 600 kA/m. Anisotropy coefficients were calculated from the hysteresis loops.

An *in situ* MFM study of the phase transformation was carried out using an atomic/magnetic force microscope equipped with a sample heating/cooling stage. The sample stage consisted of a thermoelectric cooling unit capable of varying the sample temperature from about -30°C to $+50^\circ\text{C}$, a copper transfer plate for better temperature uniformity, and a semi-enclosed chamber filled with flowing dry Argon gas to prevent condensation of water vapor at low temperatures. Each $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ sample was attached to the stage using thermal tape to ensure good thermal contact. Temperature was monitored and maintained within ± 0.2 K using a thermocouple attached to the heating/cooling stage. Each single crystal image was obtained from a different sample cut by electric discharge machining (EDM) with the specified crystal axis oriented normal to the largest sample face.

A high-purity polycrystalline sample of composition $\text{Gd}_5(\text{Si}_{2.09}\text{Ge}_{1.91})$ was also cut and polished for comparison with the single crystal samples. The domain structures during the phase transition of this sample were recorded on the same heating/cooling stage.

Single crystal transition temperatures were measured by slowly increasing and decreasing the temperature of the sample at a rate of less than 0.017 K/s while imaging the sample with both AFM and MFM. The transition temperature was recorded as the temperature at which the sample surface shifted due to the martensitic nature of the transition and magnetic contrast either appeared or disappeared, depending on transition direction. For the polycrystalline sample, the transition was recorded as the temperature at which the magnetic image began or finished changing.

2.4 CASE STUDY 4: Melt-spun $\text{Fe}_{75}\text{Si}_{10}\text{B}_{15}$ Ribbons

Fe-rich transition-metal type amorphous alloys with high boron content have been reported to have good soft magnetic properties as shown by static magnetic measurement [14]. Thus, an amorphous ribbon should provide good in-plane shape anisotropy and show significant changes in domain structure with changes in shape or stress.

As an exploratory experiment, therefore, $\text{Fe}_{75}\text{Si}_{10}\text{B}_{15}$ ribbons were melt spun and mechanically deformed in the center to produce a gradient of physical distortion through the ribbon. Two bent ribbons were then mounted for microscopy to allow for traversing the surface while maintaining a parallel plane with the surface necessary for imaging.

2.5 CASE STUDY 5: Magnetic Tunnel Junction Behavior

Magnetic tunnel junction behavior, first observed at room temperature by Moodera and co-workers [15], is a topic of great current scientific and technological interest. The electric

current through a magnetic tunnel junction relies on spin-dependent tunneling of the electrons from a ferromagnetic layer, through an insulating barrier, into another ferromagnetic layer, with the electronic spins correlated by ferromagnetic exchange forces within the magnetic domains in the ferromagnetic layers [16]. Tunnel junctions and their switching have been imaged using magnetic microscopy [17], but the true magnetic domain behavior of a spin-dependent tunnel junction as it reverses from high resistance to low resistance, and vice versa, has never before been observed while the resistance is simultaneously being measured.

The resistance of a magnetic junction depends on the relative magnetization directions in the two ferromagnetic electrodes. Together with the electron band structure – in particular the differences in the relative densities of states in the majority “spin up” half band and the minority “spin down” half band as shown in Fig. 12 [18] – the resistance of the junction will vary significantly, depending on the applied field. Parallel alignment of the magnetization in the two ferromagnetic electrodes allows the conduction electrons that tunnel through the insulating barrier layer to find a similar density of states on the other side. Conversely, antiparallel alignment of magnetic domains in the two electrodes leads to majority conduction electrons from a high density of states finding that the density of states is much lower on the other side. This results in a higher resistance in the antiparallel configuration than in the parallel configuration.

Fig. 12: Schematic of band structures in a magnetic tunnel junction showing the majority spin up and minority spin down half bands, and how the resistance is affected by the orientation of the magnetization in the electrodes on either side of the tunneling barrier [5]

Multilayers of the structure Si(substrate)-NiFe(12nm)-AlO_x(1.5nm)-FeCo(5.4nm)-CrPtMn(32.8nm)-Al(5.4nm) were fabricated into magnetic tunnel junctions by Non-Volatile Electronics, Inc. [19], as sketched in Fig. 13. The junctions of interest had a tapered ellipsoidal shape, with the full junction multilayer stack structure covering half the ellipsoidal

Fig. 13: Schematic of magnetic tunnel junction sample.

area, and the other half the exposed NiFe free layer (divided along the long axis). Enough area of the stack and free layer were exposed from beneath Al interconnects to allow imaging by MFM. Easy axes were induced perpendicular to the ellipsoid axis by annealing under an applied magnetic field. One such multilayer device, as shown in Fig. 14, was selected and connected to a circuit via wire-bonding. The tapered half-ellipsoidal shape of this junction had dimensions of $\sim 12\mu\text{m}$ at the widest point along the short axis and $\sim 40\mu\text{m}$ along the long axis. This device was then imaged on an MFM stage modified to include *in-situ* applied field capability.

During imaging, a potential of 50mV was applied across the magnetic tunnel junction. The stack was positioned in the right half of the image and the free layer in the left half for easy comparison of the junction multilayer stack to the free layer electrode. The tip of the ellipsoid from which domain nucleation began was positioned at the top of the images. As the free layer of the junction was known to have a coercivity of

$\sim 800\text{A/m}$ (10 Oe), the field was first increased to $+2.4\text{KA/m}$, decreased to -3.6KA/m , and returned to $+2.4\text{KA/m}$, with domain images taken by MFM at intervals of typically 400 A/m. Images were taken at smaller field intervals in the hysteretic region, where changes in domain structure with changing field were much more rapid, and resistances were recorded using a Keithley 3000 series sourcemeter at all field steps.

Fig. 14: Optical micrograph of NVE tunnel junction test pattern, with ellipsoid junctions visible. Magnification 250X.

3. RESULTS

3.1 CASE STUDY 1: Stress in an FeSiAl thin film resulting in stripe domains

Initial vibrating sample magnetometry measurements are summarized in figures 15, 16, and 17, with the film stresses included in figures 16 and 17.

Fig. 15: Saturation magnetization as a function of N partial pressure.

There is quite a drastic transition in properties from the 4ppN₂ film to the 5ppN₂ film evident in the stress, coercivity, and saturation field curves. Although this change is not as visible in the M_s data, the microstructural information obtained via SEM and TEM microscopy (Fig. 18) confirms a significant microstructural change in the films between those fabricated at these partial pressures of nitrogen.

Fig. 16: In-plane coercivity (H_c) and magnitude of compressive stress as functions of N partial pressure.

Fig. 17: Film saturation field and compressive stress as a function of partial pressure of N.

Fig. 18: SEM (a) and TEM (b-d) micrographs of 0ppN₂ (a and b), 5ppN₂ (c) and 10ppN₂ (d). Electron diffraction patterns are included for the TEM micrographs.

Fig. 19: Left: MFM images at remanence for 0%, 3%, 5%, and 10% pp N₂ in the sputtering gas. Right: Stripe domain width as a function of nitrogen partial pressure.

MFM results, then, were expected to show some change between the same samples. Fig. 19 shows both qualitative and quantitative differences between the domain structures at remanence. If the domain width plot is examined, it appears that there is simply an incremental change across the entire composition range. This observation is supported by the remanence images, which show both a continual decrease in domain width and apparent long range length (the lines of the stripes are less straight). However, *in-situ* applied field imagery from film to film is quite different; as can be seen in figures 20-22, the high field nucleation of domains for the higher pp N₂ samples is much more random (there is less domain pinning) and domain rotation finishes at much lower fields, corresponding to the smaller saturation fields measured in the VSM. These figures include the VSM hysteresis loops for each sample.

Fig. 20. MFM images obtained from the 0% pp N film in applied fields of (a) 19.9 kA/m, (b) 8.0 kA/m, (c) 0 kA/m, (d) -5.5 kA/m, (e) -15.9 kA/m, and (f) -20.0 kA/m

Fig. 21. MFM images obtained from the 3% pp N film in applied fields of (a) 51.8 kA/m, (b) 19.9 kA/m, (c) 0 kA/m, (d) -8.0 kA/m, (e) -43.8 kA/m, and (f) -51.8 kA/m

Fig. 22. MFM images obtained from the 5% pp N film in applied fields of (a) 19.9 kA/m, (b) 9.6 kA/m, (c) 0 kA/m, (d) -2.4 kA/m, (e) -10.0 kA/m, and (f) -19.9 kA/m

In fact, the pinning of domains despite saturation of the sample is the greatest difference seen from film to film, and is illustrated in Fig. 23. This figure shows the 0% and 5% pp N₂ films at remanence after being magnetized to saturation in opposite directions. While the 5% film shows little similarity between the two images, the 0% film demonstrates several areas that are clearly domains with complementary contrast. In effect, the domain walls have been pinned to the same location despite the apparent saturation and destruction of stripes at higher fields.

Fig. 23: MFM images obtained from the 0%pp N film at the oppositely magnetized remanent states are shown in (a) and (b). Notice the complementary contrast of the highlighted regions. The MFM images in (c) and (d) were obtained from the 5%pp N film at the oppositely magnetized remanent states

3.2 CASE STUDY 2: Magnetization reversal in CoFeHfO films

The magnetization curves along the easy and hard axes of the samples are shown in Fig. 25-26. The observed reduction in magnetization at high field in the thin CoFeHfO film (Fig. 24b) is likely due to the diamagnetic response of the Si substrate which was not subtracted

from the measured signal. The hysteresis loop parameters are summarized in Table I. The easy-axis hysteresis loops of both the thick CoFeHfO film and the thin film exhibit high squareness, suggesting that the magnetization reversal involves mostly irreversible domain

wall motion in the films. The hard-

axis loops of both films indicate

domain rotation. The anisotropy

fields were measured to be 6.0 kA/m

and 5.4 kA/m for the thick and thin

films, respectively.

Both films showed significant

changes near their coercive fields

when a field was applied along their

easy directions. Fig. 25 gives

examples of MFM images of the

thinner film under various fields

applied along the easy axis. The

domain pattern remained essentially

the same when the reversed field was

below the coercive field. As the

reversed field was increased to about

the coercive field (670 A/m), large-

Fig. 24: Hysteresis loops measured along the easy and hard directions. (a) CoFeHfO (800 nm) film, (b) CrSi/CoFeHfO (10 nm) film.

scale changes in the domain pattern were observed, accompanied by a local switching of

image contrast in regions about $0.2\ \mu\text{m}$ in size. The domain structure remained unchanged as the reversed field was further increased. Similarly, the thick CoFeHfO film exhibited greatest changes in domain structure when the reversed field was increased from about 2 kA/m to 3 kA/m, which was close to the coercivity.

TABLE I
MAGNETIC PROPERTIES OF THE THICK CoFeHfO (800 nm) SAMPLE AND
THE THIN CoFeHfO (10 nm) SAMPLE ALONG THE EASY AND HARD
AXES OF MAGNETIZATION

CoFeHfO film (thickness: 800 nm)			
	Coercivity (A/m)	Remanent magnetization (kA/m)	Susceptibility at coercive point
Easy axis	1777	794	2448
Hard axis	765	101	125
CrSi-coated CoFeHfO film (thickness: 10 nm)			
	Coercivity (A/m)	Remanent magnetization (kA/m)	Susceptibility at coercive point
Easy axis	670	785	2033
Hard axis	< 361	< 25	149

When a field was applied along the hard axis of the thin CoFeHfO film, a distinctive reversal process was observed, as shown in Fig. 26. A domain pattern (width $< 0.5\ \mu\text{m}$) with out-of-plane stray field components became apparent at about 1.3 kA/m. As the applied field was increased, the domain features rotated gradually. During this process, the image contrast *increased*, reached a maximum when the striations aligned perpendicular to the field (i.e., parallel to the easy axis) and then diminished as the sample approached saturation. This is in contrast to the results obtained from the thick CoFeHfO film that showed relatively little domain switching and a smaller change in contrast.

Fig. 26: MFM images obtained from the 10 nm thick CoFeHfO film under magnetic fields of (a) 799 A/m, (b) 2.39 kA/m, (c) 6.45 kA/m and (d) 9.95 kA/m applied along the hard axis.

Fig. 25: MFM images obtained from the 10 nm thick CoFeHfO film under magnetic fields of (a) 0 A/m, (b) 398 A/m, (c) 1.59 kA/m and (d) 2.47 kA/m applied along the easy axis. The easy axis coercivity is 670 A/m.

3.3 CASE STUDY 3: Anisotropy in a Complex Magnetic Material

Fig. 27 shows the domain structure of three different $\text{Gd}_5(\text{Si}_{1-x}\text{Ge}_x)_2$ single crystals in the ferromagnetic state, with the a, b, and c-axes perpendicular to the surface, respectively. It should be noted that each face was aligned at room temperature, so the a-axis image was slightly misaligned after a significant martensitic shift during the transition. The crystal with the c-axis perpendicular to the surface was known to change phases due to significant lateral movement of physical surface features ($\sim 15\text{ }\mu\text{m}$) at a likely transition temperature.

Fig. 27: 20x20 μm images of single crystals in ferromagnetic state (at 260K) with, from top left, a-, b-, and c-axes perpendicular to the surface.

The difference between the three axes is striking. The a-axis image exhibits stripes of relatively strong contrast and of periods of two lengths scales, the first on the scale of 1-2 μm and the second on the scale of $\sim 5 \mu\text{m}$, the b-axis highly branched rosette domains, and the c-axis shows complete uniformity. The lack of contrast in the c-axis image was verified to well below the transition temperature and for multiple transition cycles, and in all cases the transition from paramagnetic to ferromagnetic or vice versa was very rapid – within a ΔT of less than 0.1°C and a time of less than a few seconds. There was, however, a transition temperature hysteresis of $\sim 2^\circ\text{C}$ in all samples.

Fig. 28: MFM images showing the phase transition of polycrystalline $\text{Gd}_5(\text{Si}_{2.09}\text{Ge}_{1.91})$ sample (3° phase contrast, $20 \mu\text{m}$ scans). The temperature of the sample is marked for each image.

The domain structures of the polycrystalline sample were, as expected, quite different. However, there was also a marked increase in the ΔT necessary for the phase transition to progress completely. Both of these observations are illustrated in Fig. 28.

The transition temperatures measured for all samples were in good agreement with thermal expansion data, allowing for the interpretation that domain structures in the single crystals were only present in the ferromagnetic phase [20]. Fig. 29 shows the a-axis sample in the paramagnetic and ferromagnetic states as an example.

Fig. 29: a-axis single crystal $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ sample in paramagnetic and ferromagnetic states (left and right). Temperature of the sample is as marked.

Vibrating sample magnetometry data for the single crystal cube is shown in Fig. 30 and

Fig. 30: VSM hysteresis loops for a-, b-, and c-axes of single crystal $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ at 260K.

indicates quite clearly a uniaxial anisotropy along the b-axis of the crystal, with the a- and c-axes being equally hard.

In order to perform the anisotropy calculations, it is assumed that the magnetization curve is reversible and that the magnetization mechanism along the hard axis is entirely by reversible moment rotation. The anisotropy energy is then the difference in energy required to saturate magnetization along the hard and easy directions. This assumption is not entirely valid, but is often used to make analysis practical. If the energy to saturate in each direction is the area between the magnetization curve and the M axis, then the anisotropy energy is the difference in these two areas [21]. To determine the anisotropy coefficients, parametric equations were fitted to each magnetization curve and linear equations fitted to the linear portions of the a- and c-axes. For uniaxial anisotropy, $E_a = K_1 \sin^2 \theta + \dots$ [21], so

$K_I = W_{[001]} - W_{[010]} = 4.1 \pm 0.2 \times 10^4 \text{ J/m}^3$. This value for K_I is comparable to iron and to uniaxial garnets, is an order of magnitude less than Co or $\text{BaFe}_{12}\text{O}_{19}$, and is an order of magnitude larger than nickel.

3.4 CASE STUDY 4: Melt-spun $\text{Fe}_{75}\text{Si}_{10}\text{B}_{15}$ Ribbons

Images shown in Fig. 31 and 32 were arranged as a collage of separate images, spatially oriented, with each image 40 μm square. Rectangular micrographs were merged from 40 μm square single images using photo editing software.

In both Figures 31 and 32, as the strain increases (toward the right in both figures), the magnetic contrast increases, the band width of the domains increases, and the structure

transforms qualitatively. In fact, in Fig. 31c, the far right of the image shows a transition to another domain structure entirely, as highlighted below.

Fig. 31: Comparison of AFM topography (top) to MFM magnetic information (bottom). Slip planes can easily be seen in the surface, with increasing magnetic contrast and band width as the physical deformation increases.

Fig. 32: Comparison of AFM topography (top) to MFM magnetic information (bottom), similar to Fig. 13 but in a different location on the ribbon.

In this area, the most highly deformed of any of the regions, the degree and length scale of domain branching is quite high over most of the surface, even in the areas that still show regular stripes. The highlighted area appears to show a shift from the dominant stripe structure towards complete branching without the regular stripe length scale period.

3.5 CASE STUDY 5: Magnetic Tunnel Junction Behavior

For all of the following images, the junction stack is located on the right side of the image (R) and the exposed free layer is the left (L), as shown in Fig. 33. The step boundary, which is the physical edge between the junction stack and the free layer, is seen as a straight line in the center of the magnetic image, with the separation allowing for easy comparison.

Fig. 33: Representative image of junction device.

The field-resistance hysteresis loop for the junction, representative images of increasingly negative field, and increasingly positive field are shown in Fig. 34 through 36, respectively. $\Delta R/R$ for this junction was $\sim 20\%$, indicating simply that the device was working and representative of a fairly basic tunnel junction. As expected, for applied fields well outside the hysteretic region of the R-H loop, the MFM images show no magnetic contrast on either the free layer side or the multilayer stack side of the junction. This is due to saturation of the magnetization in the device plane, leaving no perpendicular contrast to image. In the hysteretic region, domain structure becomes more complex, rotating significantly out of the sample plane. This smaller scale structure appears before the resistance begins to change.

Fig. 34: Plot of junction resistance versus planar applied field.

domains nucleated in the junction at about 10 Oe in the loop of Fig. 34, and spread to the entire area through the transition and persist in the junction stack until the field strength reaches about -20 Oe, all shown in Fig. 35. Domain magnetizations return to the sample plane (two dimensional) after the field is increased beyond -20 Oe. The center magnetic flux stripe is wider and has higher contrast at negative saturation

Also of note is a possible vortex at the ellipsoid tip, and a flux closure path between the CoFe layer and NiFe free layer showing varying contrast based on the magnetization direction between the layers.

Specifically, as the field becomes more negative from positive saturation, small, three dimensional

Fig. 35: Magnetic force microscope images $8\ \mu\text{m}$ square of a section of tunnel junction, with NiFe free layer electrode on the left and junction multilayer stack on the right. Applied magnetic field strengths are, from left to right and top to bottom, 30, 9.9, 6.1, 3.7, 1.8, -2.8, -7.9, -14.1, and -42.5 Oe. Possible rotating vortex domain wall is circled.

than at positive, and almost disappears at during the transition. As the field reverses toward the positive direction, the nucleation process essentially reverses. The 3-D domains first appear in the free layer electrode and disappear last in the junction stack, and the dark flux line reappears along the physical edge at saturation. One significant difference is a larger scale domain boundary that moves toward the ellipsoid end as the field increases.

Fig. 36: Magnetic force microscope images $8\ \mu\text{m}$ square of a section of tunnel junction, with NiFe free layer electrode on the left and junction material stack on the right. Applied magnetic field strengths are, from left to right and top to bottom, -20.5, -10.5, -4.5, 1.5, 3.5, 5.5, 7.2, 11.6, and 29.5 Oe. Vortex and large-scale wall are circled.

4. DISCUSSION

4.1 CASE STUDY 1: Stress in an FeSiAl Thin Film Resulting in Striped Domains

Figure 20 shows the MFM images obtained from the same area of the 0% pp N sample at various stages of the hysteresis cycle. After the sample had been magnetized to saturation, on reducing the applied field a fine and irregular stripe domain structure nucleated (Fig. 20b). The stripe domains coarsened and became more regular as the applied field was reduced to zero (Fig. 20c). Along the steepest part of the hysteresis loop local switching of image contrast occurred, leading to connection and disconnection of the stripe domains (Figs. 20c-d). This suggests that the perpendicular magnetization component of parts of the stripe domains reversed. During this stage irreversible changes of the in-plane magnetization component also took place as indicated in the measured hysteresis loop. It was noticed that in this stage the domain width remained relatively constant and that independent switching of image contrast of parts of a stripe domain were observed. These observations seem to suggest that the irreversible changes of the in-plane component occurred mainly by local switching of domain magnetization or by local motion of short sections of domain wall. This may be accompanied by switching of the perpendicular component that was manifested as switching of the image contrast. This magnetization reversal process is different from that brought about by simultaneous motion of long domain wall sections. In the latter case, domains of the preferred magnetization direction would be observed to grow at the expense of neighboring domains as domain walls moved. Growing areas of the uniform MFM image contrast would be observed. This is contrary to the present observation that the stripe domain persisted and the domain width remained unchanged. The persistence of the stripe domain pattern could be due to the fact that the magnetostatic energy associated with it is lower than that of a

uniformly magnetized domain that has a uniform perpendicular component.

As the reverse field was increased beyond the coercive field the stripe domains disintegrated into short and irregular segments ($<0.5\ \mu\text{m}$, Fig. 20e). An interpretation of the observed change is that the process consists of local switching of domain magnetization of a few grains into the film plane (the grain size was measured to be about $0.1\ \mu\text{m}$ by TEM). This process is hysteretic as indicated in the high-field regime (from about 1.2 to 16 kA/m) of the magnetization curve. The domain width was found to decrease when the applied field was increased beyond the coercive point as a result of the disintegration of the stripe domains. Similar variations in stripe domain width with applied field in the high field regime have been observed in previous studies on other Fe-based thin films [22, 23]. Further increases of the applied field caused the image contrast to decrease as the in-plane magnetization along the field direction increased toward saturation (Fig. 20f).

Similar sequences of changes in MFM images were observed in the nitrided films. Nevertheless several differences in magnetization reversal between the 0% pp N film and the 1%–4% pp N films were noticed. For comparison the MFM images taken from the 3% pp N film are shown in Fig. 21. In the 3% pp N film a stripe domain structure was nucleated at a higher field than in the 0% pp N film (compare Fig. 21a with 20a). A larger reverse field was needed for domain switching to take place in the 3% pp N film than in the 0% pp N film, showing that domain wall pinning is stronger in the former. Similar behavior was observed in the 1, 2, and 4% pp N films. These observations are consistent with the results of the VSM measurements which show that the 1%–4% pp N films had higher coercivities than the 0% pp N film.

The domain reversal of 5% and 10% pp N films exhibited subtle differences from that observed in the 0%–4% pp N films. As shown in Fig. 22, after stripe domains were nucleated (Fig. 22b) the domain pattern of the 5% pp N film exhibited smaller changes (Figs. 22c and 22d) than in the 0%–4% pp N films along the steepest part of the hysteresis loop. As the reverse field was increased beyond the coercive field the bright stripes became wider than the dark stripes. No disintegration of the stripe domains was observed when the sample magnetization approached saturation. Similar domain reversal was found in the 10% pp N film. This observation is in contrast to that made on the 0%–4% pp N films. A possible explanation is that in the high field regime the magnetization process taking place in the 5% and 10% pp N films involved mainly uniform rotation of domain magnetization towards the sample plane, while in the 0%–4% pp N films local switching of domain magnetization occurred instead.

Evidence of strong domain wall pinning was actually observed in the 0%–4% pp N films. An example is given in Figs. 23(a) and 23(b) which shows the MFM images obtained from the 0% pp N sample at oppositely magnetized remanent states. Regions with complementary image contrast were observed. This indicates strong domain wall pinning, probably at the grain boundaries, as the domain width was of the same order of magnitude as the grain size (~ 400 nm versus ~ 250 nm, respectively). This suggestion is supported by the fact that strong domain wall pinning was not observed in the 5%–10% pp N samples. As shown in Figs. 23(c) and 23(d), the stripe domain structures found in the 5% pp N sample at the oppositely magnetized remanent states show much less repetition than in the 0% pp N sample. The observed difference in domain pinning between the two groups of films (namely the 0%–4%

films, and the 5%–10% pp N films) could be related to the change in the film structure as pp N was increased from 4% to 5%. It was found in the TEM study that the 0%–4% pp N films have large columnar bcc grains ($\sim 0.1 \mu\text{m}$), while the 5% and 10% pp N films consist of a mixture of randomly oriented equiaxed bcc nanograins (10 nm diameter or less) in an amorphous matrix (Fig. 18). Since the grains in the 5% and 10% pp N films are much smaller than the domain width, the effect of ripple and the strength of domain wall pinning are weaker than in the 0%–4% pp N films.

This data all correlates with the shapes of the VSM loops in figures 20-22, with the local switching at hysteretic field points and coherent rotation along the sloped regions of the loop, as seen in the corresponding figures and as discussed above.

4.2 CASE STUDY 2: Magnetization reversal in CoFeHfO films

As seen in Fig. 24 the easy axis hysteresis loops of both the thick CoFeHfO film and the thin coated film exhibit high squareness, suggesting by themselves that the magnetization reversal involves mostly irreversible domain wall motion across the films with very little domain rotation. The hard axis loops of both films indicate the opposite, with most of the magnetization reversal likely brought about by domain rotation. This conclusion agrees well with the domain patterns of Fig. 25 in which all observed changes were in local switching of regions about $0.2 \mu\text{m}$ in size, and to the domain patterns observed in the thicker film. Both of these changes occurred near their respective coercive fields, as would be expected. The lack of change as the field is further increased in either direction also correlates to the VSM picture of the film magnetization.

The imagery along the hard axis of the thicker film showed little either in the way of domain switching as in Fig. 25 or in changes in image contrast as in Fig. 26. Thus the magnetization reversal in this film probably involved coherent rotation of domains in the sample plane, which is usually expected in films with an in-plane uniaxial anisotropy under hard-axis applied fields. This sample can then be said to match the VSM loop predictions of domain behavior quite well. However, the drastic contrast changes and domain changes seen in the thinner CoFeHfO film (Fig. 26) are less easily explained. The domain pattern likely corresponds to the formation of magnetic ripples caused by local variations in anisotropy [24]. Adjacent domains would have anti-parallel magnetization components along the easy axis but parallel magnetization components in the applied field direction. Increasing the hard-axis field causes the domain magnetization to rotate toward the field direction in all three dimensions, leading to a maximum in contrast when the in plane components of the magnetic moments could lie along the energetically favorable easy axis. During this rotation, the contrast increases and decreases as seen in the micrographs.

4.3 CASE STUDY 3: Anisotropy in a Complex Magnetic Material

The rosette domain patterns visible in Fig. 27b and Fig. 37 are unique to a specific group of materials – ferromagnets with a relatively high, uniaxial anisotropy. While the stripes in Fig. 27a may seem to contradict this, these stripes are consistent with a small angular shift of the b-axis ($\sim 3^\circ$) as the crystal transitions from monoclinic phase to orthorhombic phase. As the samples were cut with specific axes normal to the sample plane at room temperature, the face of the ferromagnetic low temperature phase would no longer be parallel to the b-c plane of

the crystal structure. When both of these images are compared to Kerr images of a NdFeB single crystal (Fig. 37), the magnetic moment alignment of the $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ single crystals becomes quite clear, even without comparison with the VSM data. These domain structures are a result of large domains aligned along the b-axis in the bulk crystal. The domains near the surface of the crystal begin to branch into smaller domains to minimize surface stray field energy as seen in the side plane Kerr image in Fig. 37. When viewed with the easy axis of the crystal perpendicular to the surface, these branches appear as the rose patterns seen in NdFeB, cobalt, other uniaxial materials, and now this complex gadolinium-silicon-germanium material. This conclusion is in marked contrast to the a- or c-axis anisotropy expected of this material and to related Tb_5Si_4 and Tb_5Ge_4 compounds, as mentioned above. In fact, this bulk magnetic structure is counter-intuitive given the crystal structure illustrated in Fig. 11. It implies that the magnetic exchange of the Gd ion moments is not between Gd nearest neighbors in the a-c slabs as might be anticipated, but instead is indirect, propagating perpendicular and along interplanar (Si, Ge)—(Si, Ge) bonds. Despite this conceptual difficulty with the results, they are strongly substantiated by the VSM results on one single crystal in Fig. 30 and are compatible with the significant increase in interplanar Si—Ge bonding in the ferromagnetic phase from the monoclinic phase.

The peculiar patterns observed in the high purity polycrystalline samples in Fig. 28 could also be interpreted in terms of the presence of a strong magnetic anisotropy. When the easy direction is inclined at an acute angle to the surface normal, the domains in the bulk have a magnetization component normal to the surface. The branching seen in the single crystal samples would therefore occur near the polycrystalline surface as well.

Fig. 37: Side-by-side comparison of MFM images of b-axis and a-axis $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ images (top and bottom left, $20\ \mu\text{m}$ square) and Kerr images of basal and side planes of NdFeB single crystals.

As the temperature was lowered through the transition, the length scale of the domains might increase as the bulk-scale magnetic ordering increased. This is supported by thermal expansion studies at the phase transformation, which indicate a smaller range of temperature over which the transition occurs, and hence sharper order-disorder transitions in these same single crystal samples than in the polycrystalline sample [20].

4.4 CASE STUDY 4: Melt-spun $\text{Fe}_{75}\text{Si}_{10}\text{B}_{15}$ Ribbons

These ribbons also show a stripe domain structure when stressed (Fig. 31-33). When this structure is combined with the dendritic branching of the stripes (similar to the branching discussed in the case of the Gd-Si-Ge material), it is quite clear that, in this case, the tensile stress has pushed the anisotropy out of the ribbon plane. As seen in each figure, as the

degree of deformation increases, the contrast, stripe width, and stripe branching all change toward structures indicative of a strong out-of-plane anisotropy.

4.5 CASE STUDY 5: Magnetic Tunnel Junction Behavior

The line of dark contrast at the edge of the junction stack (Fig. 33, 35 and 36) indicates a strong spatial gradient of the magnetic field. The reason for such a state is that the magnetic flux path through the CoFe pinned layer either looped into the NiFe free layer (if the two dimensional configuration is antiparallel) or was repelled by the NiFe free layer (parallel configuration). This is supported by the disappearance of the line during switching of the junction and by the measured resistance of the junction device. The contrast is strongest when the antiparallel configuration drives a higher resistance and weaker when the parallel configuration drives a lower resistance. See images 1, 6, and 9 of Fig. 35 for representative examples.

Between $\pm H_C$, smaller domains with higher stray field indicate a complicated rotation mechanism for the moments in the free layer. The larger domains seen at higher fields do not rotate coherently; they nucleate, rotate independently and often out of the sample plane, and then recombine to form larger domains again in the oppositely magnetized state. These patterns are caused by the combined effects of applied magnetic field, magnetic anisotropy, and magnetostatic effects of the pointed ellipsoidal shape, the straight edge of the multilayer stack, the thin-film aspect ratio of each electrode, and the layers and their domain walls on each other. The magnetization change of the free layer is clearly not caused by simple

rotation or flipping. In fact, each reversal sequence shows a domain structure that might be a vortex (see circled regions in Fig. 35 and 36).

It should be noted that the behavior of the domains exhibited in Fig. 36 appears to match the behavior modeled for tapered ellipsoids and show similarities to “domain wall traps” modeled by McMichael et. al. [25]. The shapes are intended to nucleate domains near the center of the ellipse and allow for domain wall travel toward the ends under increasing applied field, which occurs on a longer length scale in the center three images of Fig. 36 (smaller scale domains disappear in a line from bottom to top). In the last image there is a sustained light contrast, much like the domain configuration modeled for the traps, which are of dimensions $\sim 0.5\mu\text{m} \times \sim 0.25\mu\text{m}$. Qualitatively, then, the behavior of this larger junction can serve as a model for smaller, device-sized junctions.

5. CONCLUSIONS

In the first case study involving the FeSiAl thin films, contrast and spacing of domain patterns are clearly related to microstructure and stress. As would be expected from magnetomechanical calculations, the greater the stress, the greater the perpendicular anisotropy. This is clearly seen from the increase in contrast and saturation field as the stress of the film increases (all are maximums in the 3% film). Microstructure seems to contribute most strongly to local site pinning of domains. The VSM loops for these samples can, then, be easily explained.

Case study #2 most clearly demonstrates localized, incoherent domain wall motion switching with field applied along an easy axis for a square hysteresis loop. The thicker CoFeHfO film, in fact, appeared to demonstrate expected coherent rotation behavior for the hard axis as well. However, hard axis images of the thinner film, with their unexpected perpendicular rotation behavior, demonstrate how localized imaging of domains can be almost uncorrelated to the bulk behavior of a material.

In case study #3, axis-specific images of the complex Gd-Si-Ge material clearly show the influence of uniaxial anisotropy. These MFM images qualitatively predicted the VSM measurements quite well, as proven by the anisotropy calculations performed. In effect, the images quantitatively reflect the anisotropy coefficients to within an order of magnitude, as the type of patterns seen in Fig. 27 are characteristic to materials with uniaxial anisotropy coefficient K_1 within about an order of magnitude to 10^4 J/m^3 .

Case study #4, the only study with the sole intent of creating domain structures for imaging, also demonstrated in fairly simple terms the effects of increasing stress on domain patterns. These ribbons also show a striped domain pattern when stressed, and when combined with dendritic branching of the stripes, the images again indicate that the anisotropy of the ribbon has been pushed out of the ribbon plane. Contrast and properties of stripes change with degree of deformation up to the point seen in Fig 31. The pattern seen highlighted in this figure even begins to qualitatively match the rosettes seen in the $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ single crystal.

In case study #5, it was proven that the width of magnetoresistance loops could be quantitatively predicted using only MFM. However, the magnitude of the resistance change can not be predicted, as the nature of the in-plane magnetizations of the electrode layers cannot be measured using MFM. It may be possible, then, to combine Kerr or TEM domain imaging with MFM to predict all qualities of an MR loop. The “squareness” of hysteresis was shown to be directly related to the switching of smaller domains between parallel and antiparallel states, a process that may be more characteristic of similar large junctions than smaller junctions used in memory arrays and other products. The longer scale domain behavior of this junction does, however, seem to correlate with modeling of smaller junctions, as noted in the discussion above.

When all case studies are considered together, a dominating factor seems to be that of anisotropy, both magnetocrystalline and stress induced. Any quantitative bulk measurements heavily reliant on K coefficients, such as the saturation fields for the FeSiAl films, H_C in cases 1, 3, and 5, and the uniaxial character of the $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$, transferred to and from the

domain scale quite well. *In-situ* measurements of domain rotation and switching could also be strongly correlated with bulk magnetic properties, including coercivity, M_S , and hysteresis loop shape. In most cases, the qualitative nature of the domain structures, when properly considered, matched quite well to what might have been expected from theory and calculation, and provided such information in a matter of minutes. In fact, typical characterization in each of these studies was far more complete and reliable with domain imagery to back it up – especially the single crystal and applied field pictures. In these simple cases, it appears that domain imagery may be close to standing alone in magnetic characterization. The surprises in the 10 nm CoFeHfO film, the complexity seen in the polycrystalline Gd-Si-Ge sample and the broad range predictions of the K_1 of the same reinforce the unreliability of making concrete statements based purely on domain imagery of any type, but it may be possible to create standards similar to the types used in optical microscopy for metallography in these complex cases.

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APPENDIX: COPIES OF PUBLICATIONS AND PAPERS

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The effect of nitrogen on the microstructure, stress, and magnetic properties of RF-sputtered FeSiAl(N) thin films

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Abstract

In a series of RF-sputtered soft FeSiAl(N) films, the partial pressure (pp) of N in the plasma was observed to have a profound effect on the magnetic properties, stress, and microstructure. 1% (pp) N caused the coercivity (H_c) to more than double. H_c peaked for 3% (pp) N, then decreased steeply for $N > 4\%$ (pp). Stress appears to have a major influence on the magnetic properties. Film stress correlated quite closely with H_c . Hysteresis loops appear to indicate a stripe domain structure in which the magnetization has in-plane components which are aligned parallel, but perpendicular components which alternate up and down. Stripe domains were observed directly by magnetic force microscopy. The microstructure also changed significantly with added N. 1% (pp) N caused the strong (1 1 0) texture to become very weak. For $> 4\%$ (pp) N, there was a transition from textured columnar 100 nm diameter BCC grains to a mixture of randomly-oriented, equiaxed BCC nanograins (10 nm or less) in an amorphous matrix. Transmission electron microscopy observations appear to indicate that the grain refinement and phase separation take place by a decomposition process. Saturation magnetization also decreased with added N for $> 4\%$ (pp) N, indicating that one of the two phases has a lower M_s value. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Soft magnetic materials; Sputtering, RF; Anisotropy—stress induced; Domain pattern

There has been considerable interest in sputtered FeSiAl(N) films as soft magnetic shield layers for disk drive read heads. N can have a strong effect on the microstructure and magnetic properties [1,2], yet these effects are still not well understood. In order to investigate the effect of N, a series of films was RF diode sputtered from an alloy target (85 Fe–10 Si–5 Al wt%) onto SiO₂/Si substrates using an Ar/N gas mixture, varying N partial pressure (pp), and keeping total gas pressure (9.6 mTorr), forward power (1.43 W/cm²), and film thickness (1.7 μm) constant. Magnetic properties were

characterized by vibrating sample magnetometry (VSM) and magnetic force microscopy (MFM). Film stress was determined from atomic force microscopy (AFM) measurements of the curvature of the film–substrate combination, using the equation of Eckertova [3] and the Si data of Ma [4]. Film structure and microstructure were determined from X-ray diffraction (XRD), transmission electron microscopy (TEM), and SEM fracture cross-sections. Film compositions were characterized by energy dispersive X-ray (EDX) analysis and parallel electron energy loss spectroscopy (PEELS).

Nitrogen has a strong effect on the magnetic properties of the films (Fig. 1). 1% (pp) N caused the coercivity (H_c) to more than double. H_c peaked at 3% (pp) N, then decreased steeply for $N > 4\%$ (pp). Stress appears to have a major influence on the magnetic properties. All the films showed compressive stress, the magnitude

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Fig. 1. In-plane coercivity (H_c) and magnitude of compressive stress as functions of N partial pressure.

correlating closely with H_c (Fig. 1). The in-plane hysteresis loops, while very steep near the origin, all showed curved high-field portions that suggested the presence of a stripe domain structure of the type first described by Fugiwara [5], in which neighboring stripes have aligned in-plane magnetization components, but their perpendicular components alternate up and down. MFM observations of the films at remanence showed stripe domains

in all samples (Fig. 2), with alternating light and dark contrast showing the presence of alternating perpendicular magnetization components. Stripe width (determined from Fourier analysis) decreased with increasing N content, varying from 0.51 μm for 0% (pp) N to 0.25 μm for 10% (pp) N.

The structure and microstructure also changed significantly with added N (Fig. 2). Films sputtered with 0% N showed a columnar microstructure (observed by SEM fracture cross-section), BCC crystal structure with a strong (110) texture perpendicular to the plane (from XRD and TEM), and grains approx. 100 nm in diameter (from TEM). No evidence of ordering was observed. 1% (pp) N caused the strong (110) texture to become very weak. The microstructure consisted of large columnar BCC grains out to 4% (pp) N. Between 4 and 5% (pp) N, there was a transition: films for 5 and 10% (pp) N consisted of a mixture of randomly oriented equiaxed BCC nanograins (10 nm or less diameter) in an amorphous matrix (Fig. 2). TEM observations appear to indicate that the grain refinement and phase separation take place by a decomposition process. This transition also marked a steep increase in actual N content of the films (N:Fe atomic ratio as determined by PEELS was 0.2 for 3% (pp) N, rising to 0.9 for 5% (pp) N).

Saturation magnetization (M_s) was constant from 0 out to 4% (pp) N, then decreased with added N (Fig. 3). By 10% (pp) N, it has dropped by almost 50%. This

Fig. 2. Micrographs: MFM images ($10 \times 10 \mu\text{m}$) of films (A) 0% (pp) N, (B) 3% (pp) N, (C) 5% (pp) N; (D) SEM fracture cross section of 0% (pp) N film, (E) Dark field TEM of 0% (pp) N film, (F) high resolution TEM of 5% (pp) N film.

Fig. 3. Saturation magnetization (M_s) as a function of N partial pressure.

indicates that one of the two phases (BCC nanograins or amorphous phase) has a lower M_s . It contradicts the hypothesis that N first reacts with Al until it is all reacted, then reacts with Si, then goes into the Fe lattice [1]. Were this the case, one would expect M_s to first increase with added N, as the Si and Al are 'withdrawn' from the FeSiAl film, driving it towards pure Fe composition (see data in Ref. [6]).

In summary, N greatly affects the stress, microstructure, and magnetic properties of FeSiAl(N) sputtered films. The stress and the perpendicular texture appear to set-up perpendicular anisotropy that causes stripe domains. All the films showed isotropic in-plane compressive stress, which can cause a perpendicular magnetic anisotropy component for materials with positive magnetostriction, through the magnetoelastic coupling. Compressive stress appears to adversely affect H_c . Stress magnitude and H_c variations correlate quite closely. For > 4% (pp) N, there was a microstructural transition to a two-phase mixture of BCC nanograins in an amorphous matrix. Contrary to what was found in another study [2], nanograined films as thick as 1.7 μm could be produced by this RF diode sputtering process.

In order to optimize FeSiAl(N) films for soft magnetic shield layer applications, perpendicular anisotropy should be minimized, or films should have in-plane

magnetic anisotropy, and the stripe domain structure should be avoided. Perpendicular anisotropy will itself adversely affect in-plane magnetic properties, and the stripe domains it causes can make reversal more difficult. Nanograined films (such as those produced near 5% (pp) N) should offer several significant advantages. Since the grains are much smaller than the domain sizes, there will be less pinning. There should also be much less effect of ripple (which is related to the variations in local anisotropy directions, and varies with the relative sizes of grain size and spin coupling length (see e.g. Ref. [7]). Such nanograined films have a random texture and, therefore, no magnetocrystalline contribution to perpendicular anisotropy. However, they still show enough stress-induced anisotropy to produce stripe domains, so the composition would have to be adjusted to minimize or change the sign of the magnetostriction, or alternatively process would have to be optimized to decrease compressive stress or even make a small tensile stress.

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Magnetic force microscopy study of magnetization reversal in sputtered FeSiAl(N) films

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The magnetization reversal in a series of rf-sputtered FeSiAl(N) films has been studied using magnetic force microscopy. A system has been developed which has the capability to image domain structure while an in-plane magnetic field is applied *in situ*. All films exhibited a stripe domain structure in zero applied field which was indicative of a perpendicular component of domain magnetization which alternates in sign. All films showed a similar sequence of magnetization processes: on reducing the applied field from saturation a fine stripe domain structure nucleated and then coarsened as the field was decreased to zero. Local switching of domain contrast was observed along the steepest part of the hysteresis loop as the perpendicular component reversed. As the reverse field was increased toward saturation, the stripe domains disintegrated into smaller regions. This observation is consistent with an interpretation that the domain magnetization rotated locally into the sample plane. The saturation field and the film stress exhibited similar trends with nitrogen partial pressure. The results suggest that the perpendicular anisotropy that caused the formation of the stripe domain structure could be induced by the film stress via magnetoelastic coupling.

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1. INTRODUCTION

Thin films of soft magnetic materials, such as Sendust (FeSiAl), have applications as soft magnetic shield layers in magnetoresistive recording heads. In order to improve and optimize the soft magnetic properties of these types of films it is necessary to obtain a better understanding of how the microstructure and stress affect the magnetization processes and hence the magnetic properties. In this article we report on a study of the magnetization reversal of reactive rf-diode sputtered FeSiAl(N) films, using a magnetic force microscopy (MFM) with *in situ* applied field capabilities. By directly observing the domain structure under various applied fields for films whose stress and microstructure is well characterized, we hope to obtain a better understanding of the relationship between magnetic properties, stress, and microstructure.

As-deposited FeSiAl films sputtered with Ar usually have high coercivity, low permeability and large out-of-

plane magnetic anisotropy.¹ Post-deposition annealing at elevated temperature around 400 to 600 °C is required to restore the ordered DO₃-type structure that gives soft magnetic properties.¹⁻³ Another way to achieve soft magnetic properties is to deposit Sendust films by N₂ reactive sputtering. It has been shown that addition of nitrogen to the sputtering gas can affect the grain size and stress of Fe alloy films and hence the magnetic properties.⁴ Dodd *et al.* reported that soft Sendust films up to 500 nm thick can be deposited by N₂ reactive rf magnetron sputtering, and that a substantial decrease in coercivity was found when the ratio of nitrogen flow rate to N₂+Ar flow rate was 0.2%.⁵ Nevertheless they observed an increase in coercivity and transition to columnar growth with increasing film thickness. Nanograined FeSiAl(N) films of several microns thick have been successfully prepared by dc magnetron sputtering⁶ and rf-diode sputtering⁷ using Ar+N₂ sputtering gas. In the latter study the deposited films exhibited a stripe domain structure, indicating the presence of a perpendicular anisotropy that could adversely affect the soft magnetic properties. The magneti-

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FIG. 1. MFM images obtained at the remanent state of the FeSiAl(N) films deposited with (a) 0%, (b) 3%, (c) 5%, and (d) 10% partial pressure of nitrogen in the sputtering gas.

ization reversal in these FeSiAl(N) films is the subject of the present study.

II. EXPERIMENTAL DETAILS

A series of FeSiAl(N) films of thickness $1.7 \mu\text{m}$ were deposited by rf-diode sputtering onto Si(100) substrates with 300 nm of thermal SiO_2 on the surface. Sputtering gas was an $\text{Ar}+\text{N}_2$ mixture with different partial pressures (pp) of nitrogen ranging from 0% to 10%. Hysteresis loops were measured by vibrating sample magnetometry (VSM). Film microstructure was characterized by transmission electron microscopy (TEM). Film stress was determined by measuring the curvature of the film-substrate system using an atomic force microscope. Domain structure was studied using MFM with magnetic tips that were magnetized perpendicular to the sample plane. An electromagnet capable of producing an in-plane field up to 56 kA/m (700 Oe) was built and mounted on the sample stage. During the experiment samples were first demagnetized by applying an ac field with decaying amplitude along one direction. MFM images were then taken under various fields applied *in situ*. Reproducible image contrast was obtained from the same sample in the remanent state before and after the experiment, indicating that the tip was not remagnetized by the applied field.

III. RESULTS AND DISCUSSION

In zero applied field all films showed a stripe domain structure. Figure 1 shows the MFM images of some of the samples in the remanent state. The observed stripe domain pattern and the shape of the hysteresis loops (as shown in Figs. 3–5 for the 0%, 3%, and 5% pp N films, respectively) indicate that the adjacent stripes have aligned in-plane mag-

FIG. 2. Plot of the stripe domain width as a function of the partial pressure of nitrogen in the sputtering gas.

netization component while the perpendicular magnetization components alternate in sign. This domain configuration was first proposed by Saito *et al.* who observed stripe domains in Ni-Fe films using the Bitter method.^{8,9} The alternating perpendicular magnetization component gave rise to the contrast of the MFM images since MFM measures the force gradient acting on the tip due to the perpendicular component of the surface field. In each sample the bright stripes and dark stripes were found to have similar widths, and reproducible images were obtained by rescanning the same area. These observations indicate that the domain structure of the films was not influenced substantially by the stray field from the tip. The width of the stripe domains were determined as half of the spatial period of the strongest component in the two-dimensional Fourier transform of the MFM images. As shown in Fig. 2, the domain width tends to decrease with increasing partial pressure of nitrogen in the sputtering gas during the film deposition.

Figure 3 shows the MFM images obtained from the same area of the 0% pp N sample at various stages of the hysteresis cycle. After the sample had been magnetized to saturation, on reducing the applied field a fine and irregular stripe domain structure nucleated [Fig. 3(b)]. The stripe domains coarsened and became more regular as the applied field was reduced to zero [Fig. 3(c)]. Along the steepest part of the hysteresis loop local switching of image contrast occurred, leading to connection and disconnection of the stripe domains [Figs. 3(c)–3(d)]. This suggests that the perpendicular magnetization component of parts of the stripe domains reversed. During this stage irreversible changes of the in-plane magnetization component also took place as indicated in the measured hysteresis loop. It was noticed that in this stage the domain width remained relatively constant and independent switching of image contrast of parts of a stripe domain were observed. These observations seem to suggest that the irreversible changes of the in-plane component occurred mainly by local switching of domain magnetization or by local motion of short sections of domain wall. This may be accompanied by switching the perpendicular component that was manifested as switching the image contrast. This magnetization reversal process is different from that brought about by simultaneous motion of long domain wall sections. In the latter case, domains of the preferred magnetization

FIG. 3. MFM images obtained from the 0% pp N film in applied fields of (a) 19.9 kA/m, (b) 8.0 kA/m, (c) 0 kA/m, (d) -5.5 kA/m, (e) -15.9 kA/m, and (f) -20.0 kA/m.

direction would be observed to grow at the expense of neighboring domains as domain walls moved. Growing areas of the uniform MFM image contrast would be observed. This is contrary to the present observation that the stripe domain persisted and the domain width remained unchanged. The persistence of the stripe domain pattern could be due to the fact that the magnetostatic energy associated with it is lower than that of a uniformly magnetized domain that has a uniform perpendicular component.

As the reverse field was increased beyond the coercive field the stripe domains disintegrated into short and irregular segments [$<0.5 \mu\text{m}$, Fig. 3(e)]. An interpretation of the observed change is that the process consists of local switching of domain magnetization of a few grains into the film plane (the grain size was measured to be about $0.1 \mu\text{m}$ by TEM). This process is hysteretic as indicated in the high-field regime (from about 1.2 to 16 kA/m) of the magnetization curve. The domain width was found to decrease when the applied field was increased beyond the coercive point as a result of the disintegration of the stripe domains. Similar variations in stripe domain width with applied field in the high field regime have been observed in previous studies on other Fe-based thin films.^{10,11} Further increase in the applied field caused the image contrast to decrease as the in-plane magnetization along the field direction increased toward saturation [Fig. 3(f)].

Similar sequences of changes in MFM images were observed in the nitrided films. Nevertheless several differences

FIG. 4. MFM images obtained from the 3% pp N film in applied fields of (a) 51.8 kA/m, (b) 19.9 kA/m, (c) 0 kA/m, (d) -8.0 kA/m, (e) -43.8 kA/m, and (f) -51.8 kA/m.

in magnetization reversal between the 0% pp N film and the 1%–4% pp N films were noticed. For comparison the MFM images taken from the 3% pp N film are shown in Fig. 4. In the 3% pp N film a stripe domain structure was nucleated at a higher field than in the 0% pp N film [compare Fig. 4(a) with 3(a)]. A larger reverse field was needed for domain switching to take place in the 3% pp N film than in the 0% pp N film, showing that domain wall pinning is stronger in the former. Similar behavior was observed in the 1, 2, and 4% pp N films. These observations are consistent with the results of the VSM measurements which show that the 1%–4% pp N films had higher coercivities than the 0% pp N film.

The domain reversal of 5% and 10% pp N films exhibited subtle differences from that observed in the 0%–4% pp N films. As shown in Fig. 5, after stripe domains were nucleated [Fig. 5(b)] the domain pattern of the 5% pp N film exhibited smaller changes [Figs. 5(c) and 5(d)] than in the 0%–4% pp N films along the steepest part of the hysteresis loop. As the reverse field was increased beyond the coercive field the bright stripes became wider than the dark stripes. No disintegration of the stripe domains was observed when the sample magnetization approached saturation. Similar domain reversal was found in the 10% pp N film. This observation is in contrast to that made on the 0%–4% pp N films. A possible explanation is that in the high field regime the magnetization process taking place in the 5% and 10% pp N films involved mainly uniform rotation of domain magneti-

FIG. 5. MFM images obtained from the 5% pp N film in applied fields of (a) 19.9 kA/m, (b) 9.6 kA/m, (c) 0 kA/m, (d) -2.4 kA/m, (e) -10.0 kA/m, and (f) -19.9 kA/m.

zation towards the sample plane, while in the 0%–4% pp N films local switching of domain magnetization occurred instead.

Evidence of strong domain wall pinning was actually observed in the 0%–4% pp N films. An example is given in Figs. 6(a) and 6(b) which show the MFM images obtained from the 0% pp N sample at oppositely magnetized remanent states. Regions with complementary image contrast were observed. This indicates strong domain wall pinning, probably at the grain boundaries, as the domain width was of the same order of magnitude as the grain size. This suggestion is supported by the fact that strong domain wall pinning was not observed in the 5%–10% pp N samples. As shown in Figs. 6(c) and 6(d), the stripe domain structures found in the 5% pp N sample at the oppositely magnetized remanent states show much less repetition than in the 0% pp N sample. The observed difference in domain pinning between the two groups of films (namely the 0%–4% films, and the 5%–10% pp N films) could be related to the change in the film structure as pp N was increased from 4% to 5%. It was found in the TEM study that the 0%–4% pp N films have large columnar bcc grains ($\sim 0.1 \mu\text{m}$), while the 5% and 10% pp N films consist of a mixture of randomly oriented equiaxed bcc nanograins (10 nm diameter or less) in an amorphous matrix (Fig. 7). Since the grains in the 5% and 10% pp N films are much smaller than the domain width, the effect of ripple and the strength of domain wall pinning are weaker than in the 0%–4% pp N films.

FIG. 6. MFM images obtained from the 0% pp N film at the oppositely magnetized remanent states are shown in (a) and (b). Notice the complementary contrast of the highlighted regions. The MFM images in (c) and (d) were obtained from the 5% pp N film at the oppositely magnetized remanent states.

The stripe domain structure observed in all samples indicated a perpendicular anisotropy component that could be caused by the film stress via magnetoelastic coupling. If this is the case the strength of the applied field required to magnetize the sample to saturation against the perpendicular anisotropy (i.e., the saturation field) should depend on the film stress level. In this study the saturation field H_{sat} was determined by measuring the applied field at which the MFM image contrast reduced to 15% of its starting value at zero applied field. It was found that H_{sat} first increased with pp N, attained a maximum at 3% pp N and then decreased significantly for pp N > 4% (Fig. 8). The film stress σ was found to be compressive for all samples, and the stress magnitude exhibited a trend with nitrogen partial pressure similar to that of H_{sat} as shown in Fig. 8. The close relationship between H_{sat} and σ tends to confirm that the perpendicular anisotropy is induced by the film stress caused by the presence of nitrogen.

For soft magnetic shield layer applications nanograin films, such as those produced near 5% pp N in the present study, should offer several significant advantages. The 5% pp N film has a saturation magnetization value similar to those of the 0%–4% pp N films. There is less domain pinning in the 5% pp N film since the grains are much smaller than the domain sizes. The effect of ripple, which is related to the variations in local anisotropy directions, and depends on the relative sizes of grain diameter and spin coupling length,¹² should also be much less than in the 0%–4% pp N films. Because of the random texture there is no magneto-crystalline contribution to perpendicular anisotropy in the 5% pp N film. The film has less stress and hence a lower

FIG. 7. Bright field TEM images obtained from (a) the 0%pp N film and (b) the 5%pp N film. The inset of (a) is the selected area diffraction pattern of the image. The inset of (b) is a convergent beam electron diffraction pattern showing the bcc structure of the nanograins.

saturation field (as shown in Fig. 8). However it still shows enough stress-induced perpendicular anisotropy to produce stripe domains.

Perpendicular anisotropy should be minimized as it adversely affects in-plane magnetic properties. The stripe domain structure should be avoided as it can make magnetization reversal more difficult. The film composition could be altered to give a negative magnetostriction so that an in-plane anisotropy would be induced by the compressive film stress. Alternatively the process could be optimized to decrease compressive stress or even make a small tensile stress.

IV. CONCLUSIONS

Magnetization reversal of a series of FeSiAl(N) films deposited using different partial pressures of nitrogen in the

FIG. 8. Saturation field H_{sat} and the magnitude of the compressive film stress plotted as functions of the partial pressure of nitrogen.

sputtering gas has been studied using a MFM with *in situ* applied field capabilities. Similar sequences of magnetization processes were observed in the films. All films of the series exhibit stripe domains. Substantial domain switching was found along the steepest part of the hysteresis loop. Strong domain wall pinning was observed in the 0%–4% pp N films, which have grain sizes of about $0.1 \mu\text{m}$. This was not observed in the 5% and 10% pp N films which consisted of nanograins (10 nm diameter or less) in an amorphous matrix. The saturation field was found to be closely related to the stress level indicating that the perpendicular anisotropy is caused by the film stress via magnetoelastic coupling.

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Magnetization Reversal in CoFeHfO Films

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Abstract—Magnetization reversals of a thick CoFeHfO film (800 nm), and a thin CoFeHfO (10 nm) film overcoated with CrSi (10 nm) have been studied by magnetic force microscopy (MFM) with *in situ* applied field capability. The CoFeHfO layers were deposited by rf sputtering and annealed in a magnetic field to induce an in-plane uniaxial anisotropy. For easy-axis applied fields both samples showed significant switching of MFM image contrast for fields approximately equal to the coercivity. This is consistent with the high easy-axis coercive squareness of the films. For hard-axis applied fields the thin CoFeHfO film displayed a distinctive reversal process: a ripple-like domain pattern formed and rotated gradually as the reversed field was increased. The image contrast increased, reached a maximum when the ripple-like features aligned perpendicular to the field and then diminished as the sample approached saturation. The observed magnetization reversal processes appear to be consistent with the formation of magnetization ripples which rotate toward the hard-axis applied field.

Index Terms—Amorphous and nanocrystalline materials and devices, magnetic domains, magnetic microscopy and imaging, soft magnetic materials.

I. INTRODUCTION

CoFeHfO FILMS have recently received considerable attention because of their combination of both soft magnetic properties and attractive high-frequency characteristics. It has been reported that CoFeHfO films deposited by reactive sputtering under a dc magnetic field typically have a $4\pi M_s$ value exceeding 1 T, a coercivity of a few hundred A/m and a high electrical resistivity of the order of magnitude of $10 \mu\Omega\text{m}$ [1]. These films were found to contain Fe (or Co-Fe) rich bcc nanograins and an amorphous matrix containing a large amount of Hf and O [1]. The highly resistive amorphous matrix contributes to a high resistivity of the films that significantly suppresses eddy current loss [1]. This gives rise to excellent high-frequency performance of CoFeHfO films: a reasonably high relative permeability (around 150 along the hard axis) and a low loss factor (~ 0.1) that remain constant up to hundreds of megahertz [1]. Therefore, CoFeHfO films are promising candidates for high

frequency applications including flux-guide materials in magnetic sensors or magnetic thin film inductors and converters in power supplies of telecommunication devices [2]–[4].

The aim of the present study is to obtain a better understanding of the magnetization reversal mechanism of CoFeHfO films by observing the domain structure using a magnetic force microscope (MFM) with *in situ* applied field capability. The domain structures of two CoFeHfO films with different thicknesses were observed under various applied fields, and the observations correlated with the microstructure and magnetization curves of the films.

II. EXPERIMENTAL DETAILS

A CoFeHfO film (800 nm thick) and a CoFeHfO (10 nm) film overcoated with CrSi (10 nm) were used in this study. The CoFeHfO layers of the samples were deposited by reactive rf-sputtering using an Ar + O₂ atmosphere, onto Si(100) substrates with 200 nm of SiN on the surface. These films were annealed at 250 °C for one hour in a magnetic field to induce an in-plane uniaxial anisotropy. Magnetization curves were measured from 7 mm × 7 mm samples using a vibrating sample magnetometer (VSM), along both the easy and hard axes of magnetization. Studies of the domain structure and magnetization reversals were made using a magnetic force microscope (MFM) with magnetic tips that were magnetized perpendicular to the sample plane. An electromagnet capable of producing an in-plane field up to about 56 kA/m was mounted on the sample stage. Samples were first demagnetized by applying an ac field with decaying amplitude along the desired applied field direction. Two series of MFM images were taken from each sample under various fields (up to about 43.8 kA/m) applied in situ along the easy and hard axes of magnetization of the films.

III. RESULTS AND DISCUSSION

The magnetization curves along the easy and hard axes of the samples are shown in Fig. 1. The observed reduction in magnetization at high field in the thin CoFeHfO film [Fig. 1(b)] could be due to the diamagnetic response of the Si substrate which was not subtracted from the measured signal. The hysteresis loop parameters are summarized in Table I. The easy-axis hysteresis loops of both the thick CoFeHfO film and the thin CoFeHfO film exhibit high coercive squareness, suggesting that the magnetization reversal involves mostly irreversible domain wall motion across the films. The hard-axis loops of both films indicate that the magnetization reversals in the films were probably brought about by rotation of domain magnetization. The anisotropy fields were measured to be 6.0 kA/m and 5.4 kA/m for the thick and thin CoFeHfO films, respectively.

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Fig. 1. Hysteresis loops measured along the easy and hard directions. (a) CoFeHfO (800 nm) film. (b) CrSi/CoFeHfO (10 nm) film.

TABLE I
MAGNETIC PROPERTIES OF THE THICK CoFeHfO (800 nm) SAMPLE AND THE THIN CoFeHfO (10 nm) SAMPLE ALONG THE EASY AND HARD AXES OF MAGNETIZATION

CoFeHfO film (thickness: 800 nm)			
	Coercivity (A/m)	Remnant magnetization (kA/m)	Susceptibility at coercive point
Easy axis	1777	794	2448
Hard axis	765	101	125
CrSi-coated CoFeHfO film (thickness: 10 nm)			
	Coercivity (A/m)	Remnant magnetization (kA/m)	Susceptibility at coercive point
Easy axis	670	785	2033
Hard axis	<361	<25	149

Under easy-axis applied fields the MFM images obtained from both the thick CoFeHfO and the thin CoFeHfO films showed significant changes when reversed fields approximately

Fig. 2. MFM images obtained from the 10 nm thick CoFeHfO film under magnetic fields of (a) 0 A/m, (b) 398 A/m, (c) 1.59 kA/m and (d) 2.47 kA/m applied along the easy axis. The easy axis coercivity is 670 A/m.

equal to the coercive fields of the samples were applied. An example is given in Fig. 2 which shows the MFM images of the thin CoFeHfO film under various fields applied along the easy-axis. The domain pattern remained essentially the same when the reversed field was below the coercive field [Fig. 2(a) and (b)]. As the reversed field was increased to about the coercive field (670 A/m) large-scale change in domain pattern was observed, accompanied by local switching of image contrast of regions about $0.2 \mu\text{m}$ in size. The domain structure remained unchanged as the reversed field was further increased [Fig. 2(c) and (d)]. Similarly, the thick CoFeHfO film exhibited dramatic changes in domain structure when the reversed field was increased from about 2 kA/m to 3 kA/m which is close to the coercivity of the sample. The abrupt changes in domain structure, which took place at about the coercive points in both samples, probably resulted from large-scale irreversible domain wall motions. This interpretation is consistent with the high easy-axis coercive squareness of the films as shown in Fig. 1.

When a field was applied along the hard axis of the thin CoFeHfO film, a distinctive reversal process was observed. A domain pattern (width $<0.5 \mu\text{m}$) with out-of-plane stray field components became apparent at about 1.3 kA/m [Fig. 1(b)]. As the applied field was increased, the domain features rotated gradually [Fig. 3(b) to (d)]. During this process the image contrast increased, reached a maximum when the striations aligned perpendicular to the field (i.e., parallel to the easy axis) and then diminished as the sample approached saturation. These observations indicate the presence of an out-of-plane stray field component during this magnetization reversal process. This is in contrast to the results obtained from the thick CoFeHfO film that showed relatively little domain switching (from about 4.8 kA/m to 80 kA/m) and a smaller change in image

Fig. 3. MFM images obtained from the 10 nm thick CoFeHfO film under magnetic fields of (a) 799 A/m, (b) 2.39 kA/m, (c) 6.45 kA/m and (d) 9.95 kA/m applied along the hard axis.

contrast. The magnetization reversal in the thick CoFeHfO film probably involved mainly rotation of domain magnetization in the sample plane, which is usually expected in films with an in-plane uniaxial anisotropy under hard-axis applied fields.

In the case of the thin CoFeHfO sample, the domain pattern observed under hard-axis applied fields probably corresponds to formation of magnetic ripples caused by local variations in anisotropy [5]. Adjacent domains would have anti-parallel magnetization components along the easy axis but parallel magnetization components in the applied field direction. Increasing the hard-axis field would cause the domain magnetization to rotate toward the field direction. The results of the present MFM

study indicate that during this process there was also an out-of-plane stray-field component which increased, reached a maximum and then decreased as the sample approached saturation. A possible explanation of the presence of the out-of-plane component is that the local variations in anisotropy may cause a stray field by causing changes in the local direction of magnetization.

IV. CONCLUSION

Magnetization reversals of a 800 nm thick CoFeHfO film and a 10 nm thick CoFeHfO film overcoated with 10 nm CrSi have been studied by MFM with in-plane fields applied in situ. Both samples showed significant switching of MFM image contrast for easy-axis applied fields approximately equal to the coercivity. This corresponds to the high easy-axis coercive squareness of the films. Under a reversed field applied along the hard-axis the thin CoFeHfO film displayed a domain pattern which rotated gradually as the reversed field was increased. The observed reversal processes appear to be consistent with the formation of magnetic ripples which rotate toward the applied field in the hard-axis.

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Magnetic force microscopy characterization of a first-order transition: Magnetic-martensitic phase transformation in $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$

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The magnetic-martensitic phase transformation of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($x \approx 0.5$), which occurs close to room temperature, has been observed for the first time using a magnetic force microscope (MFM) equipped with a heating-cooling stage. MFM images obtained from a polycrystalline $\text{Gd}_5(\text{Si}_{2.09}\text{Ge}_{1.91})$ sample and single crystal $\text{Gd}_5(\text{Si}_{1.95}\text{Ge}_{2.05})$ and $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ samples showed transition to a domain structure at low temperatures indicative of a ferromagnetic phase. Some samples exhibited complex domain structures, suggesting that $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($x \approx 0.5$) has a strong magnetic anisotropy. As the sample temperature increased the domain structure diminished, reflecting the transformation from ferromagnetic to paramagnetic state. On cooling the sample the domain structure reappeared, but at a lower transformation temperature than on heating. This magnetic phase transformation is highly unusual because it is an "order-disorder" phase transition, which is normally second order, but in this case the "order-disorder" (ferromagnetic to paramagnetic) transition exhibits hysteresis in temperature, indicating that it is first order. Such thermal hysteresis in phase transformation was also observed in thermal expansion experiments. The transformation temperatures obtained in the MFM study are in good agreement with those determined from thermal expansion data. © 2002 American Institute of Physics.

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I. INTRODUCTION

$\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ has recently garnered much interest due to its extraordinary response in several magnetic and electronic properties during changes in temperature and magnetic field. These include colossal magnetostriction, giant magnetoresistance, and a giant magnetocaloric effect.¹ $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ undergoes a magnetic-crystallographic transformation when $x \approx 0.5$ at a Curie temperature (typically below 270 K) which is dependent on the Si to Ge ratio. During the transformation the compound exhibits changes in strain as high as $\sim 10^4$ parts per million, magnetoresistance of about 25%, and the largest magnetocaloric effect to date.¹ The phase transition is a magnetic-martensitic transformation from a paramagnetic-monoclinic crystal structure at higher temperatures to a ferromagnetic-orthorhombic crystal structure at lower temperatures, which involves shear of sub-nanometer atomic layers in a highly complex crystal lattice through reversible breaking and reforming of covalent Si(Ge)-Si(Ge) bonds between the layers.¹

In this work the phase transitions in $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($x \approx 0.5$) have been imaged for the first time using a magnetic force microscope (MFM) equipped with a sample heating-cooling stage. The results obtained in the MFM study revealed that $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($x \approx 0.5$) in the ferromagnetic phase has a strong magnetic anisotropy, and that

the phase transition exhibits hysteresis in temperature which is unusual for an order-disorder phase transition. For comparison the thermal expansion of the samples was measured and the results confirmed the thermal hysteresis in the phase transition. The transition temperatures measured in the MFM studies and thermal expansion experiments were found to be in good agreement.

II. EXPERIMENTAL DETAILS

One polycrystalline and two single crystal samples of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($x \approx 0.5$) were fabricated from high purity elements. Their compositions are shown in Table I. The polycrystalline sample was prepared by arc-melting and the single crystal samples were grown by the Bridgman method. For the single crystal samples the b axis was in the sample plane for the $\text{Gd}_5(\text{Si}_{1.95}\text{Ge}_{2.05})$ sample but was perpendicular to the sample plane for the $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ sample. *In situ* MFM study of phase transformation was carried out using an atomic force microscope-magnetic force microscope equipped with a sample heating-cooling stage. The sample stage consists of a thermoelectric unit capable of varying the stage temperature from about 243 to 323 K. The sample was mounted on the stage using thermal tape to assure good thermal contact and the sample temperature was monitored using a thermocouple attached to the stage. The sample chamber was under positive pressure of argon to prevent condensation on the sample surface. During the experiments a sample was

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TABLE I. Transition temperatures (Kelvin). Transition temperatures and thermal hysteresis in phase transformation determined from the MFM studies and thermal expansion measurements for the polycrystalline and single crystal samples (measurement error is to 0.1 K).

Sample		On heating		On cooling		Average thermal hysteresis (K)
		MFM	Thermal expansion measurements	MFM	Thermal expansion measurements	
$Gd_5(Si_{1.95}Ge_{2.05})$	Single crystal	265.0	...	262.9	...	2.1
$Gd_5(Si_2Ge_2)$	Single crystal	270.1	269.1	267.4	267.5	2.2
$Gd_5(Si_{2.09}Ge_{1.91})$	Polycrystalline	285	288	280	281	6

cooled and heated through the transition temperatures. MFM images were taken at various temperatures which were held constant when taking the images. Phase transition temperatures were measured by scanning the sample surface continuously while slowly heating or cooling the sample and recording the sample temperature at which the magnetic image switched from a high to a low contrast or vice versa. This allowed the transition temperatures to be measured with a precision down to the accuracy of the thermocouple (0.1 K).

Thermal expansion measurements were made to study the phase transformations and to determine the transformation temperatures for comparison with the MFM results. The strains of the samples were measured using strain gages when the samples were cooled down and then heated up through a thermal cycle (between 200 to 300 K) in a closed-cycle helium refrigeration system.

III. EXPERIMENTAL RESULTS

The MFM images obtained from the polycrystalline $Gd_5(Si_{2.09}Ge_{1.91})$ sample provide conclusive evidence of thermal hysteresis in the phase transition. As shown in Fig. 1 a heavily branched domain structure was observed at low temperatures at which the sample was ferromagnetic. As the temperature increased the domains diminished and eventually the domain structure vanished at 285 K, indicating a transformation of the sample into a paramagnetic phase. The

domain structure re-appeared on cooling but at a lower transformation temperature of 280 K. Consistent transformation temperatures were measured from the thermal expansion data (Fig. 2) which also indicate thermal hysteresis occurring during the phase transformation.

MFM images of the $Gd_5(Si_{1.95}Ge_{2.05})$ and $Gd_5(Si_2Ge_2)$ single crystal samples in the paramagnetic and ferromagnetic states are shown in Figs. 3 and 4, respectively. The former shows a stripe domain structure while the latter has an irregular domain structure with a higher contrast. The single crystal samples exhibited much sharper transitions (transitions completed within 0.1 K around the transformation temperatures) than the polycrystalline sample. The single crystal samples also exhibited hysteresis in phase transformation over a temperature range of about 2 K as shown in Table I.

IV. DISCUSSION

As shown in Fig. 4, the domain pattern observed in the $Gd_5(Si_2Ge_2)$ single crystal sample is similar to those observed in highly anisotropic magnetic materials such as NdFeB and cobalt when the easy axis is nearly normal to the surface. In such cases the surface domain structure usually consists of elliptical, closed domains with a certain level of branching depending on the sample thickness.² The present MFM result seems to suggest that the $Gd_5(Si_2Ge_2)$ crystal has a strong anisotropy and the easy axis is oriented close to

FIG. 1. MFM images showing the phase transition of polycrystalline $Gd_5(Si_{2.09}Ge_{1.91})$ sample (3° phase contrast, $20 \mu m$ scans). The straight line highlighted correspond to the crystal boundary.

FIG. 2. Plot of the thermal expansion of the polycrystalline $Gd_5(Si_{2.09}Ge_{1.91})$ sample as a function of temperature over a thermal cycle.

FIG. 3. MFM phase images of the $Gd_5(Si_{1.95}Ge_{2.05})$ single crystal sample with the b axis in plane in the (a) paramagnetic and (b) ferromagnetic phase (4° contrast, $20\ \mu m$ scans).

the b axis. No other direct measurement of the magnetic anisotropy of $Gd_5(Si_xGe_{1-x})_4$ has been reported in the literature yet.

One interpretation of the stripe domain structure observed in the $Gd_5(Si_{1.95}Ge_{2.05})$ single crystal sample (Fig. 3) is that the adjacent stripes have an aligned in-plane magnetization component and out-of-plane magnetization components that alternate in sign. The latter could be lying along either the a or c axis. The a axis, along which the strain shows the largest change during the phase transition, is inclined at an angle to the surface normal. This is confirmed by $\sim 30\ \mu m$ amplitude modulations of the sample surface which were measured by monitoring the z position of the MFM probe when the sample was undergoing phase transformations. It can be deduced from the domain patterns observed in the single crystal samples that the easy axis of magnetization appears to be aligned close to the b axis and is inclined to the a and c axes. The domains in the bulk therefore have a strong magnetization component along the b axis and a component in the a - c plane. Such domain structure is consistent with the highly branched domain pattern in the $Gd_5(Si_2Ge_2)$ single crystal sample which form to minimize magnetostatic energy of the stray field emanating from the sample surface, and the stripe domains observed in the $Gd_5(Si_{1.95}Ge_{2.05})$ sample.

The peculiar patterns observed in the high purity polycrystalline samples could also be interpreted in terms of the presence of a strong magnetic anisotropy. When the easy direction is inclined at an acute angle to surface normal the domains in the bulk have a magnetization component normal to the surface. Branched domains therefore form in the near surface layer to minimize the stray field energy. As the temperature was lowered the branched domain pattern coarsened. This is in contrast to the domain patterns observed in the single crystal samples which remained unchanged. This could be related to the results obtained in the MFM and thermal expansion studies on transition width in temperature, which indicate sharper order-disorder transitions in the single crystal samples than in the polycrystalline sample. The growth of the domain pattern in the polycrystalline sample could be caused by an increase in magnetic ordering as the temperature is lowered.

The observations made in the present MFM studies have confirmed that the phase transition in $Gd_5(Si_xGe_{1-x})_4$ where

FIG. 4. MFM phase images of the $Gd_5(Si_2Ge_2)$ single crystal sample with b axis perpendicular to plane in the (a) paramagnetic and (b) ferromagnetic phase (5° contrast, $20\ \mu m$ scans).

$x \approx 0.5$ shows a thermal hysteresis, which is expected of all first-order phase transitions. The thermal expansion measurements also show this thermal hysteresis in close agreement with the MFM results. Ordinarily a magnetic order-disorder phase transition would be expected to be second order, but this is no ordinary transition. The transition is a first order magnetic-martensitic phase transition which is order-disorder (ferromagnetic to paramagnetic) in magnetism, and at the same time order-order in crystal structure (orthorhombic-monoclinic), involving a larger shear in the a axis direction.

V. CONCLUSIONS

The magnetic-martensitic phase transition of the extraordinarily responsive $Gd_5(Si_xGe_{1-x})_4$ ($x \approx 0.5$) has been characterized through magnetic imaging using a magnetic force microscope. The results indicate that the samples have a strong magnetic anisotropy in the ferromagnetic phase, and that the order-disorder transition exhibits hysteresis in temperature, a hallmark of such a first-order transition. The transition temperatures and the extent of the hysteresis were confirmed with thermal expansion data within 1 K.

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Magnetic Force Microscopy Characterization of Unusual Magnetic Coupling in an Extraordinarily Responsive Magnetic Material

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Abstract— $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ and related compounds with similar (nearly equal Si-to-Ge ratio) composition exhibit large magneto-responsive properties including a giant magnetocaloric effect, colossal magnetostriction, and giant magnetoresistance near a structural-magnetic phase transition that occurs close to ambient temperature [1]. Magnetic force microscopy (MFM) and vibrating sample magnetometry (VSM) measurements on single-crystal samples of these materials indicate that the easy magnetization axis is the b -axis of the orthorhombic magnetic phase—perpendicular to the slabs. In fact, the MFM image of a surface perpendicular to the b -axis is quite similar to domain patterns perpendicular to the easy axis of Co and other highly anisotropic magnetic materials. Therefore, it appears that $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ may require modeling similar to other multilayers and superlattices of rare-earth metals with one or more nonmagnetic constituents that exhibit long-range magnetic order across nonmagnetic layers. Many of the important phenomena of these Gd compounds could be explained by the interaction of localized Gd magnetic moments across the covalent bonding between atomic slabs, adapting models already suggested for other similar materials [2].

Index Terms—Magnetic force microscopy (MFM), magnetic materials, magnetic measurements, rare earths.

I. INTRODUCTION

IT HAS long been known that gadolinium metal exhibits a strong magnetocaloric effect, an adiabatic temperature change when magnetized, which is a function of both the magnetic entropy change and specific heat of the crystal. Due to renewed interest in the concept of magnetic refrigeration, exploratory work on Gd-based materials has uncovered several compounds that possess giant magnetocaloric effect, giant magnetostriction, and giant magnetoresistance. In $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$, all three effects are greatest at or near the phase transition temperature. The first-order, martensitic phase transition involves the lateral movement of atomic slabs with bonds created or broken between Si and Ge atoms between slabs. This movement over a distance of ~ 0.8 Å changes the crystal structure from orthorhombic (with interlayer bonding) to monoclinic (with broken interlayer bonding) and changes the magnetic structure from ferromagnetic to paramagnetic as shown in Fig. 1.

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Fig. 1. Arrangement of Gd and Si-Ge atoms, in orthorhombic (left) and monoclinic (right) phases. Differences in interplanar bonding are depicted; the a - and b -axes are in the plane of the page while the c -axis is perpendicular to the page.

This phase transition temperature is dependent on the Si-Ge ratio. In materials with an approximately equal ratio of Si to Ge, this transition occurs around room temperature with the ferromagnetic/orthorhombic state stable at lower temperatures and the paramagnetic/monoclinic state stable at high temperatures.

Since the magnetic structure of this compound was unknown, the assumption had been that the magnetic moments on the Gd atoms in the ferromagnetic state are nearly perpendicular to the b -axis, akin to magnetic structures observed in related Tb_5Si_4 and Tb_5Ge_4 compounds [3]. However, magnetic force microscopy (MFM) and vibrating sample magnetometry (VSM) measurements indicate a preferred alignment along the b -axis, with long-range magnetic coupling facilitated (or disrupted) by the presence (or lack) of interlayer Si-Ge covalent bonds.

II. EXPERIMENTAL DETAILS

In situ MFM study of the phase transformation was carried out using an atomic/magnetic-force microscope equipped with a sample heating/cooling stage. The sample stage consisted of a thermoelectric cooling unit capable of varying the sample temperature from about -30 °C to $+50$ °C. Each $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$

Fig. 2. 20 × 20-μm MFM images of single crystals at 260 K with, from top left, *a*-, *b*-, and *c*-axes perpendicular to the surface.

sample was attached to the stage using thermal tape to ensure good thermal contact. The sample temperature was monitored and maintained within ± 0.2 K using a thermocouple attached to the heating/cooling stage. Water condensation and crystallization (frost) were prevented by flowing cooled dry argon gas over the sample and platform. Each image was obtained from a different single-crystal sample that was cut by electric discharge machining (EDM) with the specified crystal axis oriented normal to the largest sample face. To determine the anisotropy of $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ vibrating sample magnetometer measurements were collected using a $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ single-crystal cube with all three axes identified using X-ray diffractometry. The sample temperature was maintained below the Curie point/transition temperature using a dry ice/ethanol mixture. Hysteresis loops were measured along the three principal axes of the crystal using a magnetizing field with a maximum amplitude of 7.5 kOe. Anisotropy coefficients were calculated from the hysteresis loops [4].

III. RESULTS AND DISCUSSION

The MFM images, each with a principal axis perpendicular to the image, are shown in Fig. 2. The rosette patterns in the *b*-axis image shown in Fig 2(b) are characteristic of images made in a plane perpendicular to the easy axis of samples with uniaxial anisotropy [5]. It appears, therefore, that the *b*-axis is the magnetic easy axis for these materials. This conclusion is further supported by the complete lack of magnetic contrast in the *c*-axis image shown in Fig. 2(c), and the stripe domain pattern of the *a*-axis image shown in Fig. 2(a). The stripe magnetic domain pattern is consistent with a significant ($\sim 3^\circ$) angular change of the *b*-axis with respect to the *a*-*c* plane from one domain to the next. After an atomic displacement in the *a*-axis direction, there

Fig. 3. VSM hysteresis loops for magnetic field applied along the crystallographic *a*-, *b*-, and *c*-axes of $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ at 265 K.

will be a misalignment of the *b*-*c* plane in the sample surface that is sufficient for the component of the *b*-axis that is out of plane to provide some perpendicular anisotropy. Surface energy is then minimized by a configuration of stripe domains, where magnetization vectors alternate between orientations with components into the plane (down) and those with components out of the plane (up).

When closely examined, the stripes seem to follow two different periods, the smaller of typically 1 μm and the larger of typically 5–8 μm. This is consistent with the smaller and larger patterns of Fig. 2(b), branches being roughly 0.5–2 μm

across and rosettes 5–10 μm across. It thus seems possible that the stripes in Fig. 2(a) could be due to components of b -axis anisotropy at an almost cross-sectional angle, similar to longitudinal, in-plane domains forced out of a film by stress. When the complete lack of magnetic contrast of Fig. 2(c) is compared to the high contrast of the other two images, it appears unlikely that any component of the spontaneous magnetization is along this axis. Therefore, the MFM results indicate that the b -axis is the easy axis, although the true easy direction could possibly include a component along the a -axis if the stripes were not due to b -axis misalignment.

The measured hysteresis loops, shown in Fig. 3(a)–(c), are consistent with those of a sample with uniaxial anisotropy. After corrections were made for demagnetizing effects along all three directions, the b -axis had the highest permeability—the magnetization curve along this direction was almost vertical at the origin—while magnetization curves along the a - and c -axes had identical permeability. The stripes seen in Fig. 2(a) but not in Fig. 2(c) may be attributed to the lattice rotation and not a component of spontaneous magnetization in the a -axis.

Anisotropy energy for magnetic moments aligned along each of the principal axes can be calculated from the area between the magnetization curve on the M – H plane, and the magnetization axis. In the case of uniaxial anisotropy, this calculation is particularly simple, as the two harder axes show the same anisotropy energy required for domain rotation under the action of a field. The anisotropy coefficient K_0 is simply the area for the easy-axis curve, while K_1 , the first anisotropy coefficient, is the difference in energy between the easy and hard axis magnetization curves [6].

IV. CALCULATION

To determine the anisotropy coefficients, exponential equations were fitted to each magnetization curve and linear equations fitted to the linear portions of the a - and c -axes. The equation or set of equations was then integrated to determine the area

between the curve and the M -axis. In this way, values of $K_0 = 0.78 \pm 0.03 \times 10^4 \text{ J/m}^3$ and $K_1 = 4.1 \pm 0.2 \times 10^4 \text{ J/m}^3$ were obtained. These values of anisotropy coefficients are similar to those of iron or a medium anisotropy garnet, and are an order of magnitude smaller than cobalt and an order of magnitude greater than nickel. The quantitative anisotropy coefficient data confirm the inferences drawn from the MFM images that the materials have uniaxial anisotropy with the b -axis as the magnetic easy axis.

V. CONCLUSION

The easy axis and anisotropy of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ have been identified using MFM and VSM. The value of $K_1 = 4.1 \times 10^4 \text{ J/m}^3$. Anisotropy is not along the directions of long-range Gd–Gd nearest neighbors—the a - and c -axes—but instead along the b -axis, indicating long-range magnetic ordering across Si–Ge nonmagnetic layers. It should be noted that the significant electronic configuration change between the two phases is the presence or absence of Si–Ge covalent bonds. Thus, magnetic order is likely to be propagated by an indirect coupling through Si–Ge bonding electrons and not through a nearest neighbor coupling between in-plane Gd moments.

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Thermal expansion studies on the unusual first order transition of $\text{Gd}_5\text{Si}_{2.09}\text{Ge}_{1.91}$: effects of purity of Gd

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Two polycrystalline samples were made by using high purity Gd and commercial Gd, respectively, but with Si and Ge starting materials of the same purity in both cases. Thermal expansion results showed that both samples exhibited a first order phase transformation, with a discontinuity in thermally-induced strain and with hysteresis in the Curie temperature. Magnetic force microscopy has been used to demonstrate the magnetic phase transformation process from paramagnetic to ferromagnetic upon cooling. It was found that the Curie temperature was lower and the thermally-induced strain higher, in the sample made from lower purity level Gd starting materials compared with the sample made from high purity Gd metal. These results indicate that the impurities (mainly C, O, N, and F) in the Gd starting material can significantly alter the strain and Curie temperature of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ alloys. © 2003 American Institute of Physics.

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Introduction

The properties of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ were first studied in 1967 (Holtzberg *et al.* 1967). In recent years, this material has attracted much attention due to its unusual giant magnetocaloric effect (MCE) (Pecharsky and Gschneidner, Jr. 1997), giant magnetoresistance (GMR), and colossal magnetoresistance (CMS) (Pecharsky and Gschneidner, Jr. 2001). It is very unusual for a single material to possess these three effects together. The magnetocaloric effect is a phenomenon on which the alignment of randomly oriented magnetic moments caused by application of an external magnetic field results in heating, while randomizing the magnetic moments by removing the magnetic field results in cooling. Previous results indicated that the impurities in starting Gd material play a critical role in determining the magnitude of the giant MCE in the final material (Gschneidner, Jr. *et al.* 2000). In this study, the effect of impurities on the magnetoelastic properties of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ system is also reported through the measurement of thermal expansion. The results show that the strain amplitude is higher in the material made from lower purity Gd.

Experimental Details

Two polycrystalline $\text{Gd}_5(\text{Si}_{2.09}\text{Ge}_{1.91})$ samples were prepared by arc-melting a stoichiometric mixture of pure components in an argon atmosphere under normal pressure. The two samples used the same purity Si and Ge starting materials (both >99.99 at.%). One of the samples was prepared using Ames Laboratory (AL) Gd(99.8% pure), the major impurities (in atomic ppm) of which were: 440-O, 200-C, 160-H. Sample 2 was fabricated by using commercial purity Gd (96.9 at.% pure) which contained 18300-O, 4300-C,

4300-N, and 3700-F (in atomic ppm). Mass losses after arc-melting were less than 0.5 wt.%, so the alloy chemical compositions were assumed to be, to a first approximation, unchanged in the final product. The thermal expansion measurements were conducted using the strain-gauge method in a Janis Research 2-stage closed cycle helium refrigeration. The sample was cooled down from room temperature through the Curie point transition, and then later was heated up through the Curie point transition. The linear thermal expansion was measured during both cooling and heating. The Curie temperatures were determined by differentiating the cooling and heating curves and finding the maximum derivative of the strain with respect to temperature. In addition, a magnetic field $B=1$ T was applied along the measurement direction to study the effect of magnetic field on the magnetoelastic properties. *In situ* magnetic force microscopy (MFM) was used to observe the phase transformation. This was carried out by using an MFM equipped with a heating-cooling stage.

Results and Discussion

Thermal expansion measurement results are shown in Fig. 1 for the sample $\text{Gd}_5(\text{Si}_{2.09}\text{Ge}_{1.91})$ made from AL Gd at $B=0$ and 1 T. An abrupt change in strain was observed at 283 K on cooling and 288 K on heating for $B=0$ T; and at 287.5 K on cooling and 298 K on heating for $B=1$ T. A discontinuity in thermal strain and thermal hysteresis are signatures of a first-order phase transition. Above the transition temperature, the sample is paramagnetic and monoclinic. Below the transition temperature, it is ferromagnetic and orthorhombic. Previous references have indicated that this first order coupled magnetic-crystallographic transition only occurs within the composition range (Pecharsky and

FIG. 1. Linear thermal strain (under fields of 0 and 1 T) of polycrystalline $Gd_5Si_{2.09}Ge_{1.91}$ made from Ames Laboratory high purity gadolinium.

Gschneidner, Jr. 1997) $0.24 \leq x \leq 0.5$. Recently, a more complete magnetic and crystallographic phase diagram has been established for the $Gd_5(Si_xGe_{1-x})_4$ system (Pecharsky et al. 2002), where a two-phase region $0.503 < x < 0.575$ is added between two regions that show a single phase around room temperature.

The composition of the present samples ($x=0.5225$) falls into this so-called two-phase region. For this composition the two crystallographic structures that can exist around room temperature are: monoclinic $Gd_5Si_2Ge_2$ -type paramagnetic state (the β phase) which is formed above 1200 K and can be retained at room temperature, and orthorhombic Gd_5Si_4 -type paramagnetic state (the γ phase) which is formed on heat treating the β phase above 690 K but below 1200 K and which appears to be stable at room temperature. When the paramagnetic β phase is cooled down below its Curie temperature, a coupled first-order magnetic-crystallographic phase transition occurs, and the ferromagnetically ordered Gd_5Si_4 -type structure (α phase) is formed. This transformation is reversible. When the paramagnetic orthorhombic Gd_5Si_4 -type γ -phase is cooled, it undergoes only an ordinary second-order transition from paramagnetic to ferromagnetic with no associated crystallographic change. This transition is also reversible. In the two-phase region, one can get either or both of β or γ phase to exist at room temperature, depending on heat treatment.

The present samples were studied in their "as-prepared" form, so that no heat treatments had been conducted on the samples. In Fig. 2, the MFM images show the transition from paramagnetic state to ferromagnetic state upon cooling. Fig. 1 shows a discontinuity in thermal strain at transition temperature of 283 K on cooling and at 288 K on heating, and hysteresis in the Curie temperature. These are both indicative of a first order phase transition. Thus taken together, they appear to indicate a first-order simultaneous magnetic-

FIG. 2. MFM images showing the magnetic phase transformation process for the sample made with high purity Ames Laboratory gadolinium. The leftmost image shows the sample in the paramagnetic state; the middle and the rightmost images show the ferromagnetic state with increasing domain size.

crystallographic phase transition. The γ phase of Gd_5Si_4 is expected to be ferromagnetic at room temperature and therefore not subject to a phase transformation upon cooling. Therefore on the basis of these results, it is believed that at room temperature, these samples adopted the crystal structure of the β phase. In addition, as shown in Fig. 1, the magnetic field can shift the Curie point to higher temperatures which also demonstrates that this first order transformation can be triggered by either temperature or applied magnetic field. In Fig. 1, the linear thermal expansion ($\Delta l/l$) of 0.26%, corresponds to a volume expansion of ($\Delta V/V = 3\Delta l/l$) about 0.78%. The thermal expansion results for sample 2 made from commercial Gd are shown in Fig. 3. On cooling $T_c = 270$ K, on heating $T_c = 275$ K. Thus the higher levels of impurities in the commercial Gd appear to have the effect of decreasing the transition temperature by $\Delta T_c = 13$ K (on either cooling or heating). The linear thermal expansion was 0.85%, corresponding to volume expansion of 2.55%. These values are considerably higher than those obtained in the sample made with high-purity Gd.

FIG. 3. Linear thermal strain in zero applied field of polycrystalline $Gd_5Si_{2.09}Ge_{1.91}$ made from commercial quality gadolinium.

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In situ applied field imaging of a magnetic tunnel junction using magnetic force microscopy

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Knowledge of domain behavior in magnetic tunnel junctions is an essential component, together with knowledge of the electron band structure, for understanding their magnetoelectronic properties. To this purpose, the magnetization reversal processes of a multilayer tunnel junction of structure substrate/NiFe/AlO_x/FeCo/CrPtMn/Al of tapered half-ellipsoid shape have been imaged using a magnetic force microscope (MFM) with *in situ* applied magnetic fields. Stripe domains through both the stack and free layers observed at zero applied field were erased by a ~100 Oe field applied to the left followed by applying a small field to the right. Magnetic domain structure did not reappear in the MFM images until a field of ~400 Oe was applied to the right. This domain pattern then persisted when the magnetic field was reduced to zero. A drastic difference in domain patterns throughout the rotational processes to saturation in each direction was also observed. When the field was applied to the left, domain walls rotated toward the direction perpendicular to the applied field before disappearing. However, in near-saturation fields to the right, domain walls formed nearly parallel to the applied field and rotated away from parallel as the applied field strength was decreased. From these images, therefore, significant insight has been gained into the magnetization processes and physical phenomena behind the magnetoresistive behavior of these junctions.

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I. INTRODUCTION

Magnetic tunnel junctions (MTJ's) that show magnetoresistance at room temperature were first observed in 1995 by Moodera and co-workers,¹ and fabricated by photolithographical techniques at NonVolatile Electronics in 1996.² MTJ's are a topic of great current scientific and engineering interest.³ The phenomena involved and the potential applications are both of significant interest. MTJ's pass a tunneling current from one ferromagnetic electrode through an insulating layer into a second ferromagnetic electrode, and this current depends on spin-dependent tunneling. The resistance of the junction depends on the relative directions of the magnetization in the two ferromagnetic electrodes as well as on the occupancy of majority and minority spin half bands. The magnetoresistive response of MTJ's depends on switching of the magnetization of the magnetic layers under an externally applied field, the details of which even now have not been fully addressed. The aim of this work is to investigate the magnetization reversal processes in MTJ's through direct observation of the domain wall processes using magnetic force microscopy (MFM) with *in situ* applied field capability.

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II. EXPERIMENTAL METHODS

The MTJ's with a multilayer structure consisting of substrate/NiFe(120)-AlO_x(15)-FeCo(54)-CrPtMn(328)-Al(54) (Fig. 1, all dimensions in angstroms) were deposited at NonVolatile Electronics on a Si wafer and patterned into a variety of devices for investigation. The junction studied was chosen for its large size (to ease MFM imaging) and its relatively large distance from other magnetic structures on the wafer (to reduce unwanted interactions and interference). It was patterned into a tapered half-ellipsoidal shape with dimensions of ~12 μm at the widest point along the short axis and ~40 μm along the long axis, with the bottom NiFe free layer a full ellipsoid. A magnetic anisotropy was induced in the NiFe layers, with the easy direction of magnetization along the short axis of the ellipsoid. A magnetic force microscope (Digital Instruments, Inc., Dimension 3100) was used in this research, employing magnetic probes (Digital Instruments, Inc., MESP) coated with CoCr. The MFM was equipped with an electromagnetic stage capable of applying variable magnetic fields up to ±600 Oe to the junction *in situ* in the sample plane. The applied field was monitored using a Hall probe embedded in the magnetizing stage. Images were taken at several constant applied fields over three-quarters of a typical hysteresis loop starting with an initial magnetiza-

FIG. 1. Schematic diagram showing the multilayered structure of the magnetic tunnel junction sample (dimensions shown in angstroms).

tion curve (zero field to high negative field to high positive field to remanence, positive being to the right of the image).

III. RESULTS AND DISCUSSION

Magnetic images of the junction when subjected to various applied fields are shown in Fig. 2 for fields applied to the left and in Fig. 3 for fields applied to the right. The junction stack is shown in the left half of each image, with the stack boundary oriented vertically down the center and the tapered tip beyond the top edge of the scan. The area to the right of the stack was the NiFe free layer electrode with its easy axis horizontal to the images, or perpendicular to the long axis of the junction multilayer.

The as-received state of the sample, as shown in Fig. 2, exhibited wide stripe domains with walls that extend straight past the stack boundary and through the NiFe electrode, with a spatial periodicity of $\sim 2.75 \mu\text{m}$. Narrow, dark contrast lines repeating over the same period were also present and extended across the physical boundary. The continuity of domains was a recurring trend throughout the image series (even after the dark lines reappeared in Fig. 3), but the stripes did not reemerge after fading, shown in the last images of both Figs. 2 and 3. These stripes were thus not likely to correspond to an equilibrium domain pattern, but more likely are an artifact of the deposition and exchange bias and magnetic easy axis processing.

The observed domain rotation and its differing behavior in opposite field directions was most unusual. As seen in Fig. 2, when the magnetic field vector pointed to the left, increas-

FIG. 2. $9.4\text{-}\mu\text{m}$ -square magnetic force microscopy images of domain configurations with junction multilayer on the left and NiFe electrode on the right. Field strengths are, from left to right and top to bottom, 0, 40, 70, 100, 40, and a return to 0 Oe, in chronological order. The arrows with single and double lines indicate the dark lines and stripe domains respectively. Junction multilayer dimensions are $\sim 12 \mu\text{m}$ in the x direction (short axis) at their widest point and $\sim 40 \mu\text{m}$ in the y direction (long axis).

FIG. 3. Magnetic force images of junction and electrode at, from left to right and top to bottom, 100, 400, 200, and a return to 0 Oe. Images are also in chronological order.

ing applied field strength resulted in a rotation of domain patterns toward the vertical axis. Increasing the field strength beyond 100 Oe did not perceptibly change the image, so the field was then reduced back toward zero. As the field was reduced, the rotation toward the vertical axis resumed, finishing at or near perpendicular to the field direction at remanence. As the field was then increased to the right (Fig. 3), all domain patterning disappeared entirely, indicating a reduction in the vertical (z) component of the magnetic moment to zero. This change is possibly due to an antiparallel moment arrangement in the pinned and free layers, providing a more energetically favorable closed flux path within the x - z plane of the sample. This lack of image contrast persisted until a field strength of 400 Oe was applied, where the same line patterns as observed before reappeared toward the tip of the ellipse. Unlike the pattern under the highest field toward the left, these lines were almost horizontal and rotated back away as the field was decreased. If the initial and final images are examined, the spacing and angle of the narrow, dark lines are found to be the same. The nominal exchange bias field at the pinned layer is 300 Oe, so the magnetization processes with the field applied to the right would appear to involve reversing magnetization at the pinned layer against its bias field, while those with the field applied to the left would be along predominantly the exchange bias field.

It is clear that the observed changes in domain pattern in this magnetic tunnel junction in response to the applied field involved a complex interaction between layer biasing, electrode easy axis, shape anisotropy of each electrode, magnetostatic interaction of the electrodes, and antiferromagnetic exchange biasing of the pinned layer. Domains are apparent in regions of the hysteresis loop where by a simple picture one would expect the pinned layer and free layer to be magnetized in the same direction. A full explanation of the differences in behavior in different applied field directions will require simultaneous measurement of both magnetoresistance and domain structure, an experiment that will be conducted subsequently. However, the interaction between the CoFe pinning and NiFe electrode layers was very clear, with magnetic domain structures continuing across the physical

edge of the junction. An antiferromagnetically coupled second CoFe layer beneath the first would likely eliminate this domain interaction by completing a closed flux path of the CoFe layers.

IV. CONCLUSIONS

Domain structures of a magnetic tunnel junction under *in situ* applied fields have been imaged using a magnetic force microscope. Coupling of the CoFe layer in the multilayer and the NiFe electrode free layer was readily observed, as was a complex domain rotation mechanism that was dependent on many factors. These factors, including field direction, stray field from the pinned CoFe layer, anisotropy of the NiFe free layer, shape anisotropy, and magnetostatic interaction between the magnetic layers, must therefore all be in-

cluded in any valid theoretical model of magnetic devices based on these types of magnetic tunnel junctions.

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STUDIES ON THE EFFECTS OF PULSED-MAGNETIC FIELD TREATMENT ON MAGNETIC MATERIALS

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Abstract. We have carried out systematic studies to investigate the effects of pulsed-magnetic field treatment on the residual stresses of carbon steels, nickel and magnetic thin film samples. A test bed was constructed for complete control of the magnetic field profile. The magnetic properties and stress states of the samples were measured before and after the treatment. Results indicate that the magnetic treatment does not have any detectable effects on the stress state of the samples.

INTRODUCTION

We have examined a proposed new nondestructive method of materials treatment that can be applied to the materials either immediately after the initial fabrication, or later after service-induced degradation has occurred. It has been claimed that this treatment can the build up of unwanted residual stress. The treatment involves application of a pulsed-magnetic field with a specific waveform to the materials. It has been claimed that such pulsed-magnetic field treatment can alter fracture resistance, wear and fatigue lifetimes, corrosion resistance, hysteretic energy losses and power conversion efficiency [1]. The implications of these claims are far reaching and it is precisely this kind of claim that has driven our research.

There have been numerous references and reports on the beneficial effects of pulsed-magnetic field treatment of metallic parts. It has been claimed that pulsed-magnetic field treatment can increase the life of metal parts by as much as 500%, with benefits found in all types of metal working from machining to grinding. Jablonowski [2] documented a large number of instances where lifetimes of tool materials have been improved as a result of pulsed-magnetic field treatment by industrial investigators of the technology. It was speculated that the increase in tool life was caused by changes in the dislocation density, residual stress, and other defects within the material such as vacancies and interstitial pairs [3]. Nevertheless these reports all appear to be anecdotal and so far there has not been any systematic study of the effects of pulsed-magnetic field treatment.

The field waveforms that have been used in pulsed-magnetic field treatment of materials consists of a low frequency component (several Hz) onto which a higher frequency component (around 100 Hz) was superimposed. Field strengths with typical amplitude of 300 to 500 Oe and cycle time of 100 to 1000 seconds are used, followed by a ten second demagnetization cycle. The effects of an ac magnetic field on material properties have been studied by Hockman *et al* [4], who claimed that application of ac fields of 300 to 2000 Oe and at frequency of 2 to 30 Hz can cause stress relief in carbon steels, high strength steels and cemented carbides. Fahmy *et al* [5] studied the effects of pulsed-magnetic field treatment on fatigue life of low-carbon steels and concluded that beneficial effects of pulsed-magnetic field treatment depend on demagnetization at the end of the treatment. It has been suggested that the pulse amplitude, pulse frequency and duration of treatment are important parameters in determining the effects of pulsed-magnetic field treatment on material properties.

The objective of this research was to determine if there were positive effects of pulsed-magnetic field treatment on material properties. Beneficial effects could include stress relief or increased component life. A further objective was to develop an understanding of how this method works. These efforts aimed to enable us to optimize the treatment conditions for its use, determine how it may best be employed and construct equipment that can be used for applying this treatment to a wide range of materials and manufactured parts. The ultimate goal was to provide a new, cleaner, faster method of stress relief that can be used in industry as an alternative to traditional stress relief annealing.

EXPERIMENTAL DETAILS AND RESULTS

This paper reports on a systematic study of the effects of magnetic processing on the magnetic and mechanical properties of several magnetic materials whose microstructures and properties were well characterized. These included (1) cold-drawn plain carbon steel (AISI 1020) bars, (2) cold-worked and annealed nickel samples and (3) magnetic thin films with high level of residual compressive stresses. A test bed consisting of pulsed-magnetic field treatment system was developed in collaboration with Technology Resource Group, Inc. for use in this work. The effects of pulsed-magnetic field treatment on the samples were evaluated by conducting nondestructive testing measurements such as eddy-current and magnetic measurements on the samples before and after the treatment. Two inspection systems, the Magneprobe and the hysteresisgraph, were used to perform the magnetic measurements. The Magneprobe allows surface magnetic property measurements to be made through the Barkhausen effect (BE). The characteristics of Barkhausen emission signal, such as the root-mean-square values and signal amplitude distribution, are dependent on the stress state of the materials. The hysteresisgraph allows bulk magnetic measurements to be made by tracing out the hysteresis curves, from which several stress-dependent parameters such as coercivity, remanence, permeability and hysteresis loss, can be obtained. X-ray diffraction (XRD) measurements were also made to detect any change in stress level in the samples. Magnetic force microscopy (MFM) was used to determine any changes in the magnetic domain structure which is closely related to the stress state of the samples.

Implementation of the pulsed-magnetic field test bed

A computer-controlled pulsed-magnetic field treatment test bed has been implemented that can be used to generate pulsed-magnetic field with specified waveforms. The field

FIGURE 1. Parameterized pulse-magnetic field signal. The input parameters are T_p , T_d , F_a , S_p and S_a that can be specified using the control software program (shown on the right) developed at the CNDE.

profile consists of a low frequency carrier signal superimposed with higher frequency components. A parameterization scheme for the pulse signals is shown in Figure 1. All the parameters can be varied, thus allowing for flexible control of the field profiles to study the effects of pulsed-magnetic field treatment on materials.

Study on cold-drawn 1020 steel bars

Cold-drawn 1020 steel bars, which possessed a high residual stress level, were used. Magnetic Barkhausen effect and hysteresis loop measurements were carried out every centimetre along the length of the samples using surface sensors before and after pulsed-magnetic field treatment.

As shown in Figure 2, a slight increase in the RMS values of the magnetic Barkhausen activity was observed after the pulsed-magnetic field treatment, indicating a possible reduction of the residual stress. The stress-relief effect of pulsed-magnetic field treatment, if there is any, is therefore very weak as indicated by the comparable magnitudes of the change in RMS BE signal voltage after pulsed-magnetic field treatment and the measurement uncertainty. We were unable to identify any significant change in the hysteresis loop properties as a result of pulse-magnetic treatment.

In order to compare the stress-relief effect of pulsed-magnetic field treatment with that of thermal anneal, a 1020 steel bar was cut into six rectangular pieces. RMS BE signal voltage were measured from the pieces with the applied field along (denoted by $V_{||}$) and perpendicular ($V_{\perp}$) to the long axis of the original steel bar. One sample was kept in the cold-worked state as a reference. Another sample was annealed at 500°C for one hour in a furnace and the other four samples were subjected to pulsed-magnetic field treatment for various periods of time. BE measurements were repeated on the samples after the thermal and magnetic treatments.

The results of the BE measurements are shown in Table 1. All the samples exhibited similar RMS BE signals in the as-received condition. The residual stress of the as-received sample was compressive along the long axis but tensile in the perpendicular direction. Thermal annealing caused $V_{||}$ to increase but $V_{\perp}$ to decrease substantially. This can be explained by the fact for plain carbon steels which have positive magnetostriction BE signal increases with tensile stress and decreases with compressive stress. Thermal annealing reduced the magnitudes of the residual stresses, causing opposite changes to the

FIGURE 2. The RMS BE signal voltage measured at various positions of a 1020 cold-drawn steel bar. The RMS Barkhausen signal increases slightly after pulsed-magnetic field treatments indicating a possible reduction in the residual stress level of the sample.

TABLE 1. Barkhausen measurements from the sample cut from a 1020 steel bar before and after various pulsed-magnetic field treatments. $V_{||}$ and $V_{\perp}$ are the measured RMS BE signal voltages with the magnetizing field applied parallel and perpendicular to the long-axis of the steel bar respectively.

Direction	Sample					
	A	B	C	D	E	F
Condition	As-received	As-received	As-received	As-received	As-received	As-received
$V_{ }$ (V)	548.6 $\pm$ 7.6	537.7 $\pm$ 4.0	527.8 $\pm$ 3.0	546.4 $\pm$ 5.0	526.9 $\pm$ 6.9	536.7 $\pm$ 2.4
$V_{\perp}$ (V)	592.2 $\pm$ 6.3	583.2 $\pm$ 4.5	564.8 $\pm$ 8.1	587.2 $\pm$ 6.2	586.0 $\pm$ 1.2	599.5 $\pm$ 6.6
Treatment	Annealed at 500°C for 1 hour	Magnetically treated for 5mins	Magnetically treated for 5mins	Kept in the as-received condition	Magnetically treated for 10mins	Magnetically treated for 30mins
$V_{ }$ (Volt)	634.3 $\pm$ 0.8	539.7 $\pm$ 3.2	543.2 $\pm$ 3.9	—	534.8 $\pm$ 1.9	533.4 $\pm$ 0.5
$V_{\perp}$ (Volt)	565.9 $\pm$ 4.2	577.8 $\pm$ 7.9	573.0 $\pm$ 2.9	—	562.1 $\pm$ 2.2	601.0 $\pm$ 2.5

BE signals in the two orthogonal directions. In contrast the samples subjected to pulsed-magnetic field treatment (i.e. samples B, C, E and F) showed much smaller changes in BE signals, indicating a much smaller effect of pulsed-magnetic field treatment on the residual stress.

Study on nickel

It was suggested that a pulsed-magnetic field may exhibit a stronger stress relief effect in nickel than in steel for two reasons: (i) nickel has higher magnetostriction constants ($\lambda_{100} = 45.9 \times 10^{-6}$, $\lambda_{111} = 24.3 \times 10^{-6}$) than iron ($\lambda_{100} = 20.7 \times 10^{-6}$, $\lambda_{111} = -21.2 \times 10^{-6}$), (ii) the shear stress exerted by a magnetic domain wall on a dislocation is 1.3 kg/mm^2 [6] which is larger than the critical resolved shear stress of nickel (0.7 kg/mm^2 at 300 K [7]). This means the interaction between the stress field of a moving domain wall and that of a dislocation may be of sufficient magnitude to move a free dislocation.

Four cold-worked nickel bars were subjected to various heat treatments (Table 2) in order to produce a series of samples with different number densities of dislocations. Magnetic domain structure of the samples was first imaged by MFM. Hysteresis loops were measured using the hysteresisgraph. Images of the domain structure in the remanent state were obtained by MFM. The sample was then subjected to pulsed-magnetic field treatment. After that MFM, hysteresis loop measurement and MFM were repeated.

In general we were unable to identify any effects of pulsed-magnetic field treatment on the properties of nickel samples. As shown in Table 3, the bulk magnetic properties such as coercivity and remanent induction were found to be unaffected by pulsed-magnetic field treatment. The MFM images of the annealed samples in general show a large scale domain structure (widths of tens of micron) which is probably related to the grain structure, and a fine scale (approximately one to two micron) pattern. The images obtained the samples after pulsed-magnetic field treatment exhibited similar features. No significant change in the domain structure of these samples was observed after pulsed-magnetic field treatment.

Study on magnetic thin films

An extensive study of the effects of pulsed-magnetic field treatment was made on FeAlSi magnetic thin films prepared by radio-frequency diode sputter deposition using 3% partial pressure (pp) nitrogen in the sputtering gas. The sample was chosen due to its high level of compressive residual stress in the as deposited state along with a well characterized microstructure. Another reason the sample was used was due to the close relationship between the sample's hysteresis loop properties and film stress which allowed us to assess the effects of pulsed-magnetic field treatment by conducting hysteresis loop measurements.

Magnetic domain structure of the samples' surface was first imaged by MFM. In-plane hysteresis loops were measured by vibrating sample magnetometry (VSM). MFM study was then repeated to image the domain structure in the remanent state. Film stress was measured by X-ray diffraction (XRD). Independent measurements of film stress were made by measuring the curvature of a long beam (along the long axis) cut from the same sample wafer. The sample was then subjected to pulsed-magnetic field treatment. After that the MFM, VSM, MFM and XRD studies were repeated to determine any change in the stress state, magnetic properties or domain structure of the sample.

The results indicated that the effect of pulsed-magnetic field treatment on the sample, if there is any, is below the sensitivities of the techniques used in the present study. The position and the width of the α -Fe peaks measured from the XRD spectra were found to remain unchanged within measurement uncertainties after pulsed-magnetic field treatment as shown in Table 4. This suggests that the lattice strain and hence the residual stress level of the sample did not change after pulsed-magnetic field treatment. It was found in the curvature measurements that the film stress was compressive, with stress levels estimated at 278 MPa before pulsed-magnetic field treatment and 290 MPa after pulsed-magnetic

TABLE 2. Sample preparation and heat treatment conditions

Sample	Treatment
1	Kept in the as-received cold-worked condition.
2	Annealed in vacuum at 400°C for 1 hour and then furnace-cooled.
3	Annealed in vacuum at 700°C for 1 hour and then furnace-cooled.
4	Annealed in vacuum at 1000°C for 1 hour and then furnace-cooled.

TABLE 3. Magnetic hysteresis properties of the nickel samples before and after pulsed-magnetic field treatment.

Sample	Treatment	Coercivity (Oe)		Remanence (G)	
		Before pulsed-magnetic field treatment	After pulsed-magnetic field treatment	Before pulsed-magnetic field treatment	After pulsed-magnetic field treatment
1	As-received	13.6	14.0	2311.1	2509.4
2	Annealed at 400°C	13.6	14.9	2491.6	2544.8
3	Annealed at 700°C	7.7	7.7	2688.3	2746.3
4	Annealed at 1000°C	2.8	3.3	1143.5	2032.5

TABLE 4. Positions and widths of the α -Fe peaks in the XRD spectra obtained before and after pulsed-magnetic field treatment.

θ Peak	Before pulsed-magnetic field treatment		After pulsed-magnetic field treatment	
	Peak position	Peak width	Peak position	Peak width
Fe (110)	44.03°	0.63°	44.03°	0.62°
Fe (211)	81.69°	0.96°	81.74°	1.08°
Fe (220)	97.81°	1.70°	97.81°	1.71°

field treatment. The result also indicates that the film stress remained unchanged after pulsed-magnetic field treatment.

The hysteresis loops obtained in the VSM measurements before and after pulsed-magnetic field treatment, as shown in Fig. 3, are essentially the same. Hysteresis loop properties such as coercive force, remanent magnetization and maximum magnetic susceptibility remained unchanged after pulsed-magnetic field treatment. These properties are known from previous studies to be stress dependent. The hysteresis loops obtained both before and after pulsed-magnetic field treatment shows curved high-field portion ($50 \text{ Oe} < H < 300 \text{ Oe}$) which corresponds to rotation of the out-of-plane domain magnetization component towards the sample plane. The observed loop shape suggests that after pulsed-magnetic field treatment the sample still maintained a strong perpendicular anisotropy induced by the compressive film stress.

FIGURE 3. Hysteresis loop of the magnetic thin film sample measured by VSM before and after pulsed-magnetic field treatment.

Results obtained from the MFM study show different domain structures depending on the magnetic history of the sample. Nevertheless the effects of pulsed-magnetic field treatment can still be evaluated by comparing the MFM images obtained before and after pulsed-magnetic field treatment at the same magnetic state (e.g. at remanence). An example is given in Figure 4 which shows strikingly similar domain patterns in the remanent state before and after pulsed-magnetic field treatment. This indicates the presence of strong pinning sites for magnetic domain walls. These pinning sites are probably the amorphous secondary phase formed at the grain boundaries as observed in the TEM studies, or other structural defects such as highly dense dislocation clusters. The present result shows that pulsed-magnetic field treatment was unable to heal these microstructural defects.

CONCLUSIONS

Detailed and systematic studies were made to investigate the effects of pulsed-magnetic field treatment on the residual stresses of carbon steels, nickel and magnetic thin film samples. A test bed was constructed that allows complete control of the applied field profile. The effects of the magnetic treatment were evaluated by measuring the magnetic properties and stress states of the samples before and after the treatment. Results indicate that the stress-relief effect of pulsed-magnetic field treatment on the samples, if there is any, is much weaker than claimed in the previous studies. The present investigation, using a range of characterization methods on a number of different specimens, both bulk samples and thin films and with a variety of chemical compositions, was unable to detect any effect.

FIGURE 4. MFM images showing the magnetic domain structure of the same area of the magnetic thin film sample in the remanent state (a) before, and (b) after pulsed-magnetic field treatment. Note the strikingly similar domain patterns (e.g. the regions circled).

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MAGNETIC FIELD GRADIENT MEASUREMENT ON MAGNETIC CARDS USING MAGNETIC FORCE MICROSCOPY

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Abstract. The magnetic field gradients of magnetic stripe cards, which are developed for classifying magnetic particles used in magnetic particle inspections, have been measured using a magnetic force microscope (MFM). The magnetic force exerted on a MFM probe by the stray field emanating from the card was measured to determine the field gradients. The results are in good agreement with the field gradients estimated from the magnetizing field strengths used in the encoding process.

INTRODUCTION

Magnetic particle inspection (MPI) is widely used as a rapid, convenient method for detecting surface or sub-surface cracks in ferromagnetic materials. Indication of defects by magnetic particles requires sufficiently strong leakage field at the defects, which is usually achieved by magnetizing the sample under test. To obtain the best inspection results, the sensitivity of the magnetic particles to defects needs to be adjusted primarily through establishing a proper magnetization level in the sample [1]. Magnetic stripe cards have recently been developed as a tool for establishing the level of the stray field gradient required for particle indication at defects for a given type of magnetic powders. These cards are similar to credit cards, ID cards and "electronic" keys in that they consist of a magnetic stripe magnetized (encoded) to give a specific magnetic pattern. The magnetic stripe cards used for MPI have different zones of periodic magnetization patterns encoded into the stripe using various magnetizing field strengths. Each of these zones is specified by a particular stray field gradient.

Sensitivity of a given type of magnetic particle can be ascertained by spraying the particles to the card and finding the zone of the lowest field gradient to which the particles adhere. The usefulness of magnetic stripe cards as a quantitative tool requires prerequisite knowledge of the stray magnetic field gradients above the stripe surface, which can be obtained using a magnetic force microscope (MFM). By this method a magnetic probe was moved towards the surface of the magnetic stripe and the force acting on the probe, which is proportional to the product of the magnetic field gradient and the magnetic moment of

the probe, was measured. In this study the magnetic field gradients of different zones of a magnetic stripe card sample were determined using the MFM technique. For comparison, the sensitivity of commercial magnetic particles was checked using the stripe card sample and a test ring (SRM 1853 from the National Institute of Standards and Technology (NIST)) magnetized using a central current carrying conductor.

MAGNETIC FORCE MICROSCOPY AND FORCE CURVE MEASUREMENTS

Magnetic force microscope (MFM) has proved a powerful tool for imaging magnetic domain structure of materials because of its high sensitivity and spatial resolution (typically 50 nm or better). The technique is an offspring of atomic force microscopy. It employs a sharp magnetic tip (the typical tip radius is about 20 to 40 nm) attached to a flexible cantilever to sense the magnetic interaction between the tip and a sample. Magnetic force as small as 10^{-11} N acting on the tip can be measured by shining a laser beam on the cantilever and detecting the reflected beam using a split photo-detector to monitor the cantilever deflection. A magnetic image of the material can be obtained by mapping the tip-sample interaction as a function of the tip's height when it is being scanned over the material surface.

In addition to imaging magnetic structure, MFM can also be used to measure quantitatively the stray field gradient above the surface of magnetic materials. In general the z-component (normal to sample surface) of the magnetic force F_z exerting on the MFM tip by a stray magnetic field H is given by [2]:

$$F_z = \mu_0 \int_V \left(m_x \cdot \frac{\partial H_x}{\partial z} + m_y \cdot \frac{\partial H_y}{\partial z} + m_z \cdot \frac{\partial H_z}{\partial z} \right) dV, \quad (1)$$

where m_i and $\partial H_i / \partial z$ ($i = x, y$ and z) are the components of the magnetic moment of the MFM tip and the stray field gradients respectively. In the situation where the length of the MFM tip is much smaller than the spatial period of the stray field pattern, one can assume that the tip moment is localized at a singular point within the tip (i.e. the tip is considered as a magnetic point-dipole) [3]. This point-dipole approximation was used in the present study to estimate the stray field gradient. Its use can be justified by the fact that the height of the MFM tip is about 10 to 15 μm , while the spatial period of the domain pattern encoded into the stripe card is about 127 μm . Since the MFM tip was magnetized along the z-direction, Equation (1) can be simplified into

$$F_z = m_z \cdot \frac{\partial H_z}{\partial z} \quad (2)$$

The z-component of the stray field gradient $\partial H_z / \partial z$ can therefore be determined by measuring the magnetic force acting on the tip. In this study force-distance curves, which is a plot of the magnetic force as a function of the vertical position of the tip above the stripe surface, were measured to estimate the stray field gradients above the stripe card surface using Equation (2).

EXPERIMENTAL DETAILS

The magnetic stripe card sample used in this study was provided by Circle System, Inc. The magnetic stripe on the card sample was magnetized by means of a sinusoidal encoding process as illustrated in Fig. 1. During the encoding process the magnetic stripe was passed under an electromagnet at a constant speed. A magnetizing coil energized by an ac current (60 Hz) was used to generate a reversing field which magnetized the stripe to give a periodic magnetization pattern. The spatial period of the pattern is determined by the frequency of the current and the speed of the card motion, while the stray magnetic field gradient of the magnetization pattern is varied by controlling the current amplitude and hence the magnetizing field strength. Eleven zones of periodic magnetic patterns with the same spatial period d ($= 127 \mu\text{m}$) but different stray field gradients were encoded using various magnetizing field strengths (Table 1). The stray fields H of the zones were measured by the manufacturer of the magnetic stripe card. The stray field gradients were calculated using $2H/d$, and the results are shown in Table 1.

Force-distance curve were measured from different zones of the magnetic stripe card using a magnetic force microscope operated in tapping mode. All measurements were made using the same MFM probe (a silicon pyramidal tip coated with a CoCr thin film) to allow direct comparison of results obtained from different zones. Before measuring a force curve a MFM image of the magnetic pattern was obtained. An example is shown in Fig. 2 (a). The bright and dark contrast represents different regions above which the MFM tip sensed a repulsive and an attractive force respectively. In order to compare with the field gradient data provided with the card manufacturer, force-distance curve measurement was made at the point where the field gradient is a maximum. The MFM signal level, which corresponds to the phase shift of the oscillating cantilever due to the tip-sample interaction, is proportional to the second derivative of the stray field gradient. The point of maximum stray field gradient, where the MFM image signal level is equal to zero, was then located from the line scan of the MFM image (as illustrated in the trace of Fig. 2 (b)) for force-distance curve measurements. During the measurements an oscillating MFM tip was moved towards and away from the surface of the magnetic stripe in a cyclic manner. The dc component of the photo-detector signal, which corresponds to amount of bending of the cantilever under the magnetic interaction between the tip and sample, was measured as a function of the vertical position of the tip above the stripe surface. A force-distance curve was then obtained by converting the photo-detector signal into force signal. The magnetic force at a fixed height above the stripe surface was simply read off from the curve, and the stray field gradient was calculated using Equation (2).

For comparison the sensitivity of magnetic particles was checked by observing magnetic particle indication on the stripe card sample and on a standard test ring (SRM 1853-124 test ring) obtained from NIST. To ensure repeatability of the test, the test ring was first magnetized to saturation by applying a dc current of 2300 A to a conductor through the center of the ring and the current was then reduced to zero. During the tests dc currents ranging from 1000 A to 2000 A were used to magnetize the ring. Magnetic particles were applied to the ring which was then examined for magnetic particle indication near the holes on the test ring. The stray magnetic field gradient A_t (in $\text{A}\cdot\text{m}^{-2}$) at ring surface was estimated using $A_t = A_n \times I_t / 1500$, where I_t is the applied current (in A) and A_n is the certified value of field gradient (in $\text{A}\cdot\text{m}^{-2}$) of the n -th hole of the test ring at a current rating of 1500 A [4]. The sensitivity of the magnetic particles were determined by identifying the hole with the lowest field gradients where a magnetic particle indication was still observed.

FIGURE 1. Schematic diagram illustrating the process of encoding a magnetization pattern into the magnetic stripe.

FIGURE 2. (a) MFM image (phase image) of one of the zones of the magnetic stripe card. (b) Line-scan of the MFM image showing the signal variation across the image. The arrow indicates the point of maximum field gradient where the force-distance curve measurement was made.

RESULTS AND DISCUSSION

A typical force-distance curve measured from the stripe card is shown in Figure 3. The curve can be divided into five sections regarding the tip-sample interaction. When the MFM tip is approaching the stripe surface from a long distance the cantilever's deflection (bending of the cantilever) is small (region A). When the tip is in close proximity of the sample surface the cantilever suddenly "snaps" into contact with the stripe because of an attractive tip-sample interaction (e.g. van der Waals or capillary forces) [5]. This is manifested as a dip in the force-distance curve (region B). The maximum forward deflection of the cantilever multiplied by the cantilever's spring constant gives the maximum attractive force on the tip. As the cantilever is being lowered it remains in contact with the stripe and is pushed back through the zero force position. Further

FIGURE 3. A typical graph showing the photo-detector signal measured as a function of the tip's position above the strip surface. The figures on the right illustrates the tip-sample interaction in different stages of the force-distance curve measurement.

lowering the cantilever bends the cantilever upward elastically, resulting in what is known as the constant compliance regime (region C) in which the cantilever's deflection is a linear function of its vertical displacement. The slope of the straight line is useful as it provides a calibration of the photo-detector signal against the cantilever's deflection. On retracting the cantilever it remains adhered to the sample surface (region D) until it suddenly breaks free of the attractive force and snaps back to the undeflected state (region E).

In order to measure the stray field gradients, the measured photo-detector signal as a function of the tip's vertical displacement was converted into a force-distance curve as follows. The photo-detector signal was first converted to cantilever's deflection using the slope of the constant compliance regime. The points of zero deflection, where the tip experienced zero force, were located by examining all the force-distance curves. The force exerting on the MFM tip was calculated by multiplying the cantilever's deflection by the cantilever's spring constant, which was estimated to be $0.5 \text{ N}\cdot\text{m}^{-1}$ from the measured resonant frequency of the cantilever using the formula derived by Sader *et al* [6].

The force-distance curves obtained from different zones of the stripe card are shown in Fig. 4. All the force curves exhibit similar features such as sudden jump of the MFM tip into contact and gentle pull-off of the tip from the stripe surface, which indicate the presence of a long-range, attractive interaction between the tip and the sample. It is evident that the jump-to-contact (i.e. attractive force) is larger and the pull-off is more gentle for the zones encoded using stronger magnetizing fields. This is attributed to the fact that these zones have stronger stray magnetic fields (and hence higher field gradient), and therefore they exerted a stronger magnetic force on the tip in addition to the van der Waals and capillary forces. Such effect is manifested as stronger attraction of the tip when it was approaching the sample surface and stronger adhesion when it was being retracted from the sample surface.

FIGURE 4. Force-distance curves measured from (a) zone 1, (a) zone 3, (a) zone 5 and (a) zone 9 of the magnetic stripe card.

The stray field gradients of different zones of the stripe card were determined from the tip-sample interactions measured at a fixed height of 500 nm above the stripe surface. This height was chosen because it has been shown that when there is no long-range magnetic or electrostatic interaction between the tip and sample, the cantilever should sense no force at a tip-sample separation of several hundred nanometers or above [7]. A value of $m_z = 6 \times 10^{-3} \text{ A}\cdot\text{m}^2$ was assumed for the MFM tip used in this study [8]. The estimated values of the stray magnetic field gradients are shown in Table 1 for different zones of the stripe card. The field gradients estimated using a MFM technique were found in good agreement with those measured by the card manufacturer. Typical error of the field gradient measured by the MFM technique is $\pm 15\%$, which comes mainly from the uncertainties in the values of the spring constant and magnetic moment of the MFM tip.

The leakage field gradients estimated using the test ring are shown in Table 2. Magnetic particle indication was observed for field gradient of about $1.5 \times 10^6 \text{ A}\cdot\text{m}^{-2}$ and greater. This is in contrast to the results of the present study, because particle indication was not observed on zone number 11 of the magnetic stripe card where the field gradient was estimated to be about $1.7 \times 10^8 \text{ A}\cdot\text{m}^{-2}$ using the MFM technique. The discrepancy could be due to the way a Ketos test ring was calibrated. The measurements involved the use of a Hall sensor of finite size (1.5 mm wide and 2.75 mm long) at a certain liftoff (about 0.1 mm) to measure the leakage field. This tends to give a field gradient smaller than the

TABLE 2. Stray field gradients ($\times 10^6 \text{ A}\cdot\text{m}^{-2}$) estimated for the holes of a test ring (SRM 1853-124 test ring) using the procedures suggested by NIST. Field gradients marked with an asterisk correspond to the case in which the magnetic particle indication was marginally observed, and the field gradients in shaded boxes correspond to cases where magnetic particle indication was not observed.

Hole	1000 A	1100 A	1200 A	1300 A	1400 A	1500 A	1600 A	1700 A	1800 A	1900 A	2000 A
1	11.3	12.3	13.4	14.5	15.6	16.7	17.8	18.9	20.0	21.2	22.3
2	5.4	5.9	6.4	7.0	8.1	8.1	8.6	9.1	9.7	10.2	10.7
3	2.7	3.0	3.3	3.6	4.1	4.1	4.4	4.6	4.9	5.2	5.5
4	1.4*	1.6	1.7	1.9	2.1	2.1	2.3	2.4	2.6	2.7	2.9
5	0.7	0.8	0.9	1.0	1.1	1.1	1.2	1.3	1.3*	1.4	1.5*
6	0.3	0.4	0.4	0.4	0.5	0.5	0.5	0.6	0.6	0.6	0.7
7	0.3	0.3	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.5	0.5
8	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.3	0.3	0.4	0.4

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TABLE 1. Comparison between the magnetic field gradients measured using a MFM and the data provided by the manufacturer of the magnetic stripe card.

Zone	Field gradient measured by card manufacturer ($\text{A}\cdot\text{m}^{-2}$)	Field gradient determined by force curve measurement ($\text{A}\cdot\text{m}^{-2}$)
Zone 1	1.28×10^9	1.24×10^9
Zone 2	1.21×10^9	1.20×10^9
Zone 3	1.14×10^9	1.14×10^9
Zone 4	1.05×10^9	1.05×10^9
Zone 5	9.59×10^8	1.04×10^9
Zone 6	8.52×10^8	8.66×10^8
Zone 7	7.28×10^8	7.78×10^8
Zone 8	6.39×10^8	6.36×10^8
Zone 9	4.79×10^8	4.62×10^8
Zone 10	3.55×10^8	4.04×10^8
Zone 11	1.95×10^8	1.68×10^8

actual value. This argument has been supported by theoretical modeling of the leakage field using the finite element method [9]. The modelling results show that the peak value of the field gradient above the surface of a Ketos ring for an applied current of 1500 A is $9.5 \times 10^7 \text{ A}\cdot\text{m}^{-2}$, which is roughly six times larger than the estimated value ($1.5 \times 10^6 \text{ A}\cdot\text{m}^{-2}$) shown in Table 2.

CONCLUSIONS

Quantitative measurements of magnetic field gradient on magnetic cards, which are being developed as a quantitative tool for classifying magnetic particles used in magnetic particle inspections, have been made using a magnetic force microscope (MFM). The field gradient was determined by measuring the magnetic force acting on a magnetic probe, which is proportional to the product of the gradient of the magnetic field emanating from the card and the magnetic moment of the probe. The results were found to agree well with the field gradients estimated from the magnetizing field strength used in encoding the magnetic patterns in the cards.

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