MATERIALS REPORTS

Technical reports giving an overview of progress and challenges in areas of materials research will be included in this section periodically.

Surface, interface, and thin-film magnetism

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A comprehensive review and state of the art in the field of surface, interface, and thin-film magnetism is presented. New growth techniques which produce atomically engineered novel materials, special characterization techniques to measure magnetic properties of low-dimensional systems, and computational advances which allow large complex calculations have together stimulated the current activity in this field and opened new opportunities for research. The current status and issues in the area of material growth techniques and physical properties, characterization methods, and theoretical methods and ideas are reviewed. A fundamental understanding of surface, interface, and thin-film magnetism is of importance to many applications in magnetics technology, which is also surveyed. Questions of fundamental and technological interest that offer opportunities for exciting future research are identified.

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I. INTRODUCTION

With the information revolution and the ever-growing need to acquire, store, and retrieve information, the science and technologies attached to magnetic recording have experienced an explosive growth. Central to those pursuits is the materials science of magnetism as it applies to surfaces, interfaces, and thin films.

Magnetism is an electronically driven phenomenon, weak compared with electrostatic effects but subtle in its many manifestations. It is quantum-mechanical in nature, with its origins in the Pauli exclusion principle and the existence of the electron spin. It leads, nonetheless, to a large variety of short- and long-range forces, and both classical and quantum-mechanical effects. This last feature provides the richness of textures and properties encountered in magnetic systems, from which useful engineering and technical applications arise.

The preparation and especially the control of surfaces and interfaces in magnetic systems open a new area in the science of magnetism, one that involves a highly interdisciplinary endeavor: physics, chemistry, and materials sciences; theory and experiment; surface science; small laboratory and central-facility research; materials preparation and characterization; academic, national-laboratory, and industrial research.

The present report is the result of the deliberations of a Panel convened in Santa Fe, New Mexico, on June 18–21, 1989, under the auspices of the Department of Energy, Council on Materials Science. The Panel, of twelve members, was chaired by Falicov and Pierce. The Panel's charge was to assess the state of the art in the area, identify the major, important issues, and estimate the prospects for future research.

Several technical developments are responsible for the intense activity in the field. In addition to the application-driven pressures mentioned above, three major advances are to be noted:

(1) The advent of new sample-preparation techniques which now permit the manufacture of single-purpose devices to extraordinarily accurate specifications; these techniques [Molecular Beam Epitaxy (MBE), Metal-Organic Chemical Vapor Deposition (MOCVD), sputtering, lithography, etc.] are becoming increasingly available and less expensive and have engendered, in addition to the obvious technological progress, a new branch of "pure" science concerned with artificially made systems.

(2) The availability of better and sophisticated sample characterization techniques, based mostly (although not exclusively) on centrally located facilities. These techniques are based on x-ray and ultra-violet photons (synchrotron sources), visible and infrared photons (ordinary and free-electron lasers), neutrons (reactors and pulsed neutron sources), and electrons of a variety of energies (electron microscopes of several kinds; low-, intermediate-, and high-energy electron sources for elastic and inelastic scattering experiments). To these should be added the existence and ready availability of excellent controlled environments (good vacuum and clean gaseous atmospheres; from very low to very high temperatures; high and spatially uniform magnetic fields).

(3) The increasing availability of fast, operationally inexpensive and numerically intensive computers which have permitted the calculation of a large variety of problems related to realistic systems, in complicated geometries, with subtle quantum-mechanical effects, and/or for practical devices.

This combination of factors makes it almost a necessity to evaluate, even though briefly, where the field is, where it is going, where the needs are greater, and where the better pay-offs may lie.

II. THEORETICAL BACKGROUND

Theoretical techniques relevant to understanding magnetic phenomena at surfaces, interfaces, and in thin films are grouped into five general areas: electronic-structure techniques, phenomenological and model system theories, theories of critical phenomena, transport theory, and the special phenomenology of micromagnetics.

A. Electronic structure

Electronic-structure techniques compute the ground state of a many-electron solid at zero temperature. A hierarchy of techniques exists in which successively more terms in the Hamiltonian are approximated from physical models or experimental data. Current *ab initio* techniques require only the specification of atomic positions and species to determine the ground-state energy. They typically use the local density approximation to the density functional formalism. With state-of-the-art supercomputers, calculations can be performed for up to ten-atom ordered unit cells and for free surfaces and interfaces. The following magnetic properties have been or can be calculated:

(1) Because magnetic energies are much smaller than binding energies, it is necessary to solve the structural problem from the outset. Physical structures may be optimized by comparing the total energies for a series of atomic configurations. Bulk lattice parameters are predicted[1] generally to within 1%, and elastic constants to within 10%. It should be noted, however, that the bulk lattice constants predicted for the magnetic 3d transition metals[2] are too small by as much as 3%. Physical structure determinations for thin films, surfaces, and interfaces are straightforward but extremely time-consuming because of the reduced symmetry. Early calculations for thin films and surfaces did not allow for

interlayer relaxation; recent calculations, as discussed below, indicate that such relaxations can significantly affect the computed magnetic properties. More complicated surface reconstructions remain to be explored.

(2) Magnetic moments of bulk transition metals and some ordered alloys are typically calculated[3] to within 2%. Rare-earth ions can be treated by these techniques only if the *f*-shell configurations are properly constrained.[4] Calculations of moments at surfaces, interfaces, and in few-monolayer films, if the positions of the atoms are correctly specified, can be expected to have the same accuracy as the bulk moments. In particular, the magnetic moments of surfaces and few-monolayer films have been predicted to be significantly enhanced[5] and, for some normally nonmagnetic materials, surface layers are predicted to acquire a magnetic moment.[6] However, these calculations have of necessity assumed atomic spacings close to the bulk values; i.e., structural relaxations have not been included. A calculation for Fe/W, allowing surface layer spacings to relax to their minimum energy configuration, shows a nearly total disappearance of the enhanced magnetic moment.[7]

(3) Magnetic structures can be predicted by comparing total energies for a limited set of magnetic structures (which exclude any spin canting) calculable by these techniques. For example, the possible antiferromagnetic phases of bulk manganese have been calculated.[8] In general such calculations agree with experimental results, with the notable exception that an antiferromagnetic face-centered cubic phase is erroneously predicted by the most accurate calculational techniques as the stable phase of iron.[9]

(4) Calculated Fermi surfaces of magnetic metals show good agreement with experiment[10] in some cases (Fe), not as good[11–13] for others (Co, Ni). Reliable Fermi surfaces are necessary for predicting transport properties.

Systematic studies of a wide variety of physical and magnetic structures of surfaces and interfaces currently require more approximate methods of electronic structure calculations. If such methods are constructed to reproduce known experimental or *ab initio* results, predictions can be expected to be quite reliable.[14]

Ab initio methods based on the local density approximation replace real electron correlation potentials and energies by average values from a homogenous electron gas, thus effectively giving a one-electron description. For certain systems in which electron correlations are important in determining the magnetism, an understanding of the electronic structure may be obtained only through explicit many-body techniques.

New techniques which combine molecular dynamics simulations with *ab initio* electronic structure determinations are still in their infancy,[15] but may be expected, in the long term, to be applied to realistic magnetic systems to determine the physical and magnetic structures simultaneously.

B. Phenomenology and model systems

There are several properties of magnetic materials which, although derived from the electronic structure, are not adequately treated by current electronic-structure techniques; these frequently omit the relativistic spin-orbit coupling terms. (Spin-orbit coupling terms have been included in *ab initio* electronic structure calculations, but usually only for closed shell, i.e., nonmagnetic systems.) Spin-orbit related energies are usually several orders of magnitude smaller than those associated with changes in physical or magnetic structure. These properties, which include anisotropy, magnetostriction, and magneto-optic coupling, have historically been treated by phenomenological models in which the *form* of the required terms in the Hamiltonian is constrained by symmetry and the magnitude of the relevant coefficients is extracted from physical models and experimental results.

Magnetic anisotropy is the energy associated with a specific orientation of the magnetic moment relative to the crystal axes or macroscopic structure. Work on itinerant electron systems, where the anisotropy derives from the entire Fermi surface, relies on empirical models. (Early calculations deriving anisotropy from tight-binding band structures, including the spin-orbit term as a perturbation, were in only fair agreement with experiment.[16]) For rare-earths and transition-metal oxides, where the anisotropy is associated with local moments, crystal-field methods have been successful; they, however, almost invariably include some adjustable parameters.[17] Empirical models for anisotropies at surfaces were developed long ago,[18] but even today they require experimental parameters.

Magnetostriction is a change in shape of a body on the application of a magnetic field. Linear magnetostriction is the coupling between the direction of the moment and elastic strain; i.e., it is the strain derivative of the anisotropy energy. Models for magnetostriction at surfaces are intimately related to surface anisotropy, but have not received much attention. The mismatch strain at an interface has a large effect on magnetic properties through magnetostriction.

Magneto-optical effects arise from the coupling between the spin and charge polarization, again a spin-orbit effect. Magneto-optical coefficients have usually been derived from experimental results.

Extension of phenomenological theories of spin-orbit related properties to surfaces and interfaces usually requires the inclusion of lower symmetry terms in the model Hamiltonians. Temperature dependence can

be included in these theories by making, phenomenologically, the coefficients depend on temperature. Model theories have been developed which relate the temperature dependences of anisotropy and magnetostriction to the temperature dependence of the moment; they are reasonably successful for local-moment systems.

C. Critical phenomena theories

Surface, interface, and thin-film magnetism provide a fertile ground to explore critical phenomena, in particular those that arise in response to restricted dimensionality, finite-size effects, and surface-driven mechanisms. It is well known that, in two dimensions, systems with one degree of freedom (i.e., Ising-like) have a well-defined phase transition.[19] In fact, the thermodynamics of that problem, solved exactly by Onsager more than 35 years ago, is one of the landmarks in the Theory of Phase Transitions. It predicts, for the simple square lattice, a transition temperature T_C equal, in energy units, to 2.269 times the value of the nearest-neighbor Ising exchange parameter. It also finds[19–22] that, as the temperature T approaches T_C from below, the magnetization of the system decays to zero as

$$M = M_0[1 - (T/T_C)]^{0.125}.$$

In contrast, isotropic systems with two or three degrees of freedom (i.e., xy- and Heisenberg-like) exhibit no long-range order in two dimensions[21–23] at any finite temperature T. The development of three-dimensional order as such systems are built up layer by layer has also been studied.[24]

There are, in addition, fascinating surface effects related to a variety of critical phenomena: behavior and transitions involving the decay in short-range order[23] (the so-called Kosterlitz-Thouless transition), the interplay between surface and bulk effects[25] (including the persistence of order on the surface at temperatures higher than the bulk Curie or Néel temperatures and various temperature dependences of the magnetization of the surface layers as compared to the bulk), and distinction between universal and nonuniversal behavior of magnetic overlayer systems when the coverage is fractional.[26]

D. Transport properties in magnetic systems

The study of transport properties in magnetic systems differs from that in any other material by the fact that it always takes place in the presence of an intrinsic, local magnetic field; in other words, it is always the study of galvanomagnetic properties—in particular, magnetoresistance.

When a magnetic field is applied to a normal (i.e., not ferromagnetic) metal, the resistance is seen to *increase* with the intensity of the field, regardless of the relative orientation of the field with respect to the current and with respect to the crystallographic axes. This phenomenon, known as ordinary or *positive magnetoresistance,* is very well understood, and for high-purity metals with a large electronic mean-free path, yields accurate and easily interpretable information about the electronic structure, the Fermi surface in particular, of the metal.[27] Increases in resistance of many orders of magnitude (a factor of a million is fairly common) are observed in particularly pure, single crystals at very low (liquid helium) temperatures and high magnetic fields (typically 10 to 100 kOe). For polycrystalline samples and at normal temperatures more modest increases, typically of a factor of 2 to 10, are obtained for equivalent fields. Positive magnetoresistance can be interpreted, in general terms, by noting that in the presence of a magnetic field, electron trajectories become convoluted (e.g., helical), and the effective distance that an electron can transport charge before being scattered decreases as the magnetic field increases.

In ferromagnetic systems, which in the absence of an applied field consist of several magnetic domains, the phenomenon of *negative magnetoresistance*[28] is observed: the application of an external magnetic field decreases the resistance by up to an order of magnitude in fields as small as 100 Oe. The phenomenon is commonly interpreted based on the fact that the external field changes the domain structure and produces a single-domain crystal. Under those conditions two effects take place. The electron trajectories, because of the presence of a now uniform internal field, become less convoluted, and the removal of the Bloch walls eliminates a source of electron scattering.[29] Both effects result in longer mean-free paths upon application of a magnetic field, i.e. a *negative magnetoresistance.*

E. Micromagnetic theory

Micromagnetic theory provides a framework for predicting macroscopic magnetic phenomena, such as domain walls and hysteresis loops, in systems where the details of the atomic structure are not important.[30] Input to the calculations includes exchange parameters (typically taken from spin-wave dispersion data), crystalline anisotropy constants (typically taken from torque curves), and sample microstructure (typically taken from electron micrographs). It is a classical (i.e., non quantum-mechanical) many-body problem in which much of the computational expense comes from the long-range nature of the magnetostatic interaction. The memory dependence of the problem means that the motion of the magnetization should be traced in time to ensure accuracy: description by the Landau-Lifshitz-Gilbert equations appears to be adequate in this respect. The theory has spawned numerous calculations which, while usually only semi-quantitative in nature,

have provided considerable insight. Quantitative accuracy is frequently prohibited by the need to include the effects of thermal fluctuations and/or a precise domain nucleation mechanism as precipitated by defects. Fortunately, materials exist in which a nucleated domain can be assumed to exist, and the major question in hysteresis is whether the domain can pass some barrier. This approach led to a domain-wall pinning theory[31] which predicted the approximate scaling of the coercive force with material parameters and to quantitatively accurate predictions for hysteresis loops in CoNi thin films.[32] Quantitative accuracy for domain walls in soft materials (where details of microstructure and other complications may frequently be discarded) has been achieved by several workers.[33]

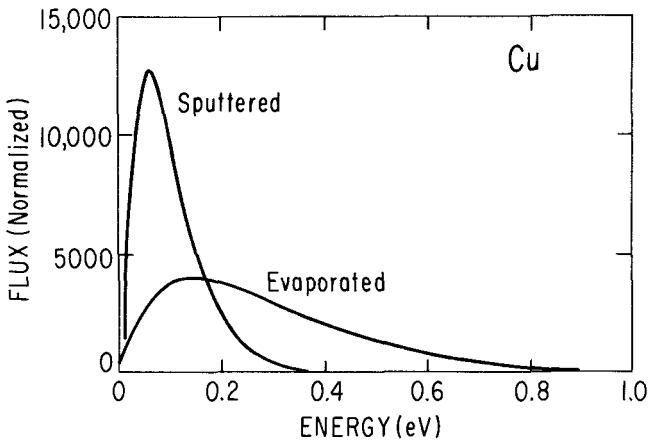

FIG. 1. Energy distribution of particle flux arriving at a substrate for sputtering at a pressure of 10 mT and substrate-to-target distance of 6 cm, and for thermal evaporation.[34]

III. MATERIALS

A. Growth techniques

In this section some commonly used preparation techniques are described. The most extensively used techniques for the growth of modern magnetic materials are sputtering and Molecular Beam Epitaxy (MBE). Both techniques have produced high-quality samples, when grown under appropriate conditions.

1. Sputtering

In sputtering the target material is bombarded with a beam of inert gas ions (ordinarily argon) and the sputtered atoms are collected on a temperature controlled substrate. Sputtering using magnetically confined plasmas is ordinarily denoted by magnetron sputtering. The pressure and substrate-target distance control the energy distribution of the particles arriving at the substrate. Under appropriate conditions[34] it is possible to deposit particles with an effective energy approaching the sputtering-gas kinetic energy (~200 °C). Figure 1 shows a comparison of the energy of Cu atoms arriving at a substrate under typical evaporation conditions and under the sputtering conditions specified in the caption. Note that under these conditions, evaporated atoms exhibit a high energy tail and are centered at much higher energies than sputtered atoms. This fact, of course, implies that under high sputtering pressures and large substrate target distances sputtering produces less damage than thermal evaporation. An additional advantage of sputtering is that the energy distribution of particles can be tuned to higher energies by decreasing the pressure and/or the substrate-to-target distance. However, contamination due to the inert gas and the presence of impurities such as oxygen makes sputtering inappropriate for the growth of semiconductors where the presence of small amounts of impurities is known to affect severely the physical properties. Moreover, because of the presence of the inert gas it is not customary to use *in situ* characterization techniques. Sputtering is the growth technique of choice in industrial applications where large-area homogeneous films are required at reasonable cost. Laser ablation is another related, more recent entry in the arsenal of the thin-film fabrication methods. In this technique a target close to the exact (or near exact) stoichiometry of the final films is bombarded ("ablated") by a laser beam, to move the material from the target to the final film. The method is particularly well suited to those materials which have widely different sputtering rates. It has been very successfully used for the growth of high-temperature superconducting oxides.[35]

2. Molecular beam epitaxy (MBE)

In MBE a number of particle beams are prepared by thermal evaporation from Knudsen cells or electron-beam guns in ultrahigh vacuum (UHV), typically of 10^{-11} Torr. The evaporation rates are kept slow and controlled using quartz crystal monitors, optical detection methods, or mass spectrometers. Control of the substrate temperature and growth rate is essential if smooth ledge growth is to be achieved, with sharp interfaces and minimal interdiffusion. It is customary to use *in situ* characterization tools such as Reflection High Energy Electron Diffraction (RHEED) and intensity oscillation of the elastically reflected or diffracted electron beams. The interpretation of the RHEED intensities and diffraction patterns has undergone a considerable evolution, although to date this technique is not on the same quantitative footing as x-ray or neutron diffraction techniques (see Sec. IV, Techniques and Facilities).

3. Metal-organic chemical vapor deposition (MOCVD)

A variety of other techniques have also been used to grow magnetic materials, especially oxides. However, they have not been used as extensively as MBE and sputtering. MOCVD is a technique by which metal atoms are carried by a large easily dissociated organic molecule from a source container to the substrate. On striking the substrate, the molecule momentarily sticks, but can be readily dissociated either by maintaining the substrate at a high temperature or by irradiating it with sufficiently-high-energy light. Common molecular carriers are the metal carbonyls. Using iron pentacarbonyl, excellent single-crystal films of Fe have been grown epitaxially on GaAs at 175 °C substrate temperature. Post-growth analysis exhibited normal bulk magnetic properties and showed no evidence of entrapped carbon.[36] Plasma-assisted MOCVD takes advantage of the added parametric control of the composition through the use of a "plasma" in the deposition chamber. Fine tuning of the final composition occurs by adjusting both the plasma and the source conditions for the desired result.

4. Production techniques

State-of-the-art systems typically might use dc-magnetron sputtering techniques with targets comparable to sample or sample pallet in size (a few inches in diameter for single samples to a couple of feet across for pallet systems). Deposition rates are kept high in large-volume products, if possible, in order to utilize effectively an expensive machine. Metals are commonly deposited at rates as great as 200 Å/s, at base pressures approaching 10^{-7} Torr. Substrates for different materials may require either heating or cooling. Amorphous magneto-optic media on plastic substrates must obviously not get hot. On the other hand, some crystal structures, grain sizes, and material phases require substrate temperatures of a few hundred degrees Celsius (100–400 °C for magnetic media). Typically, multiple layers must be deposited, so it is not uncommon to have in-line deposition systems. This fact requires complicated mechanical-transport systems if continuous processing, as opposed to batch processing, is to be used. Large volumes of deposited materials tend to flake off the insides of vacuum chambers and create defects; hence most in-line systems are oriented and deposit material horizontally or with the substrate facing down.

Simple Chemical Vapor Deposition methods can be used to deposit a variety of oxides.[37] The method essentially involves holding an appropriate single-crystal substrate close to a sublimating metal-halide source in the presence of a pressure of about 15 mm of water vapor. Typical substrate temperatures are 700 °C, with bromides used as cation. In this fashion thicknesses up to about ~20 μm can be readily achieved.

Other processes may use electron-beam evaporation if very high rates are demanded for extended periods of time. An example of this might be continuous coating of a flexible substrate for videotape. In this case maintaining material composition from the melt may become an issue and methods of monitoring composition, and replenishing depleting alloy elements are required.

B. Growth modes

Generally it is hoped that the growth of a perfect, defect free, flat, completely segregated film can be achieved over large macroscopic areas of the substrate. In practice, the growth of films proceeds by a variety of so-called "growth modes".[38] These have been traditionally categorized in the epitaxial literature as the layer-by-layer, layer-and-island, and island growth modes. The particular growth mode depends on the relative binding energies of the overlayer-overlayer atoms and the overlayer-substrate atoms. In the case of heteroepitaxial growth there are two limits: close to matching of atomic structures of the overlayer and substrate ("lattice matching") and completely different structures and atomic radii ("lattice mismatched") systems. A heteroepitaxial system which is close to lattice matching in many cases will slightly strain (if the energetics permits it) to match the substrate ("pseudomorphic growth"). This strain can be partially relieved by the formation of dislocations in thin films. Strained-layer properties and dislocation formation are current topics of investigation in semiconductor heterostructures, but have not been fully addressed for the metallic systems that are the main topic of this report. Despite strain relief caused by plastic deformation at the growth temperature, a film remains clamped to the substrate during subsequent measurements at reduced temperatures. These epitaxial constraints can exert profound effects on the magnetic phase diagram and on the general behavior of a particular phase.[39] For systems that exhibit large lattice mismatches the substrate can predetermine the relative epitaxial orientations and even the structure of the overlayer through mechanisms which are not well understood. Although a variety of epitaxial systems has been grown over many years,[40] many systems have been prepared under uncontrolled conditions or poor vacuum, making it unclear how much of the pre-existing literature is relevant for issues concerning growth and structure at the atomic level. Because structural and magnetism issues are intimately related to each other, the understanding of the magnetism strongly depends on a detailed understanding of the structural properties.

Chemical and structural disorder at growth interfaces are important in the overall issue of structure and

magnetism. Structural disorder (roughness) can increase or decrease with growing thickness, and chemical disorder depends on a variety of growth parameters such as substrate temperature, growth rate, energy of deposited particles, etc. Although growth is a nonequilibrium phenomenon and therefore kinetically controlled, thermodynamic phase diagrams are often found to play a dominant role.[40] The main reason is that surface diffusion may suffice to cause interdiffusion at a level of 2 to 5 atomic planes at an interface between two materials that form continuous sets of solid solutions. Indeed, materials that are lattice matched and have the same crystal structure frequently form solid solutions in their thermodynamic phase diagram.[41] All these considerations point to the fact that it is very important to characterize properly the materials after they are grown by a number of *in situ* and *ex situ* techniques (see Sec. IV, Techniques and Facilities). The physical properties may even reflect structural features which are not readily detected by purely structural probes. One obvious example is the dependence of the electrical resistivity on layer thickness in superlattices.[42] It is generally found that the electrical resistivity scales inversely with the layer thickness, indicating the presence of a large amount of electronic scattering at seemingly perfect interfaces.

C. Systems highlights

A large number of different systems have been grown by the techniques described above (see Tables I, II, and III). These include single epitaxial films of almost all magnetic elements, including transition metals and rare earths. Many of these elements have been grown in ultrahigh vacuum down to submonolayer thicknesses.

TABLE I. Lattice constant (in Å) of representative substrate/film combinations for some magnetic metal films (for further details see Ref. 72).

Substrate		Film	
fcc			fcc
		Ni	(3.62)
Cu	(3.61)	Co	(3.55)
		Fe	(3.59)
fcc			bcc
NaCl	(5.64)		
AlAs	(5.62)	Fe	(2.867)
GaAs	(5.65)		× 2
Ge	(5.66)		
ZnSe	(5.67)		5.733
fcc			bcc
LiF	(4.02)		
Al	(4.05)	Fe	(2.867)
Au	(4.07)		$\times\sqrt{2}$
MgO	(4.31)		
NaF	(4.62)		4.054

TABLE II. Superlattice systems (for further details see Ref. 42, p. 139).

System	Preparation method	System	Preparation method
Ni/Cu	Ev, SpDC	Fe/Mg	Ev
Ni/Mo	SpDC	Fe/V	Ev
Ni/Cr	Ev	Fe/W	SpDC
Ni/C	Ev	Fe/Ta	SpDC
Ni/V	SpDC	Fe/Y	Ev
NiFe/TiN	SpDC	Fe/Pd	Ev, SpDC
Co/Cu	SpRF, Ev	Fe/Cr	Ev
Co/Au	SpRF	Fe/Mn	Ev
Co/Nb	Ev	Fe/FeO	Sp
Co/Sb	Ev	Fe/Nd	SpDC
Co/P	El	Fe/Gd	Ev
Co/Pd	SpRF, Ev	Fe/Tb	Sp
Co/Cr	Ev	FeB/Ag	SpDC
Co/Mn	Ev	FeCo/Si	Sp
Co/Gd	SpDC	FeCo/Tb	Ev
CoNb/CoTi	Sp	Mn/Ag	Ev
CoSiBi/CoTi	Sp	Mn/Sb	Ev
Fe/Cu	SpRF, Sp	Dy/Y	Ev, MBE
Fe/Ag	Ev	Er/Y	MBE
Fe/Au	SpRF	Gd/Y	Ev, MBE
Fe/Sb	Ev	Tm/Lu	Sp
Fe/Sn	Ev		

Ev = Evaporation
Sp = Sputtering
El = Electrolytic method
MBE = Molecular Beam Epitaxy

TABLE III. Metal-on-metal growth (for further details see Ref. 73).

	Overlayer						
Substrate	V	Cr	Mn	Fe	Co	Ni	Rare earth
Cu(100)		X		X	X	X	
Cu(110)				X			
Cu(111)				X	X	X	
Ag(100)	X		X	X		X	
Au(100)		X		X			
Pd(100)			X	X			
Pd(111)				X			
Ru(0001)			X	X			
Ru(10$\bar{1}$0)				X			
Re(0001)						X	
W(110)				X			Eu Gd Tb
V(110)							Ce Dy
Fe(100)						X	
Ni(100)							Gd
CuAu(111)				X			
$Cu_3Au(100)$				X			
Y(0001)							Dy Er Gd Ho

1. Surface and monolayer films

While it is possible for theorists to model ideal monolayers in computer simulations, it has proven an almost insurmountable challenge for experimentalists to grow idealized model systems in the laboratory. The issue is associated with the need for a substrate and the inability to realize free-standing monolayers. Interactions with the substrate invariably dominate most properties of interest. For instance, Cu, Ag, and Au single crystals are good substrate candidates because of their filled *d* bands. But it is this very characteristic that creates metallurgical problems: their lower surface free energies, compared to that of the magnetic elements, can provide thermodynamic driving forces for surface segregation, intermixing, etc. For the Fe/Cu(100) system it is known that intermixing at the interface prevents the realization of monolayer structures.[43,44] Elevated substrate temperatures during growth of multilayer thicknesses of Fe on Cu(100) have been shown to provide an intermixed buffer layer that separates the pure Fe and pure Cu regions; this buffer layer stabilizes the antiferromagnetic phase[45] of fcc Fe. On the other hand, the relatively discrete interface formed by low growth temperatures yields a ferromagnetic fcc phase of Fe.[46,47] This is a clear example of the influence of growth conditions on the properties of the resultant film.

Transition-metal substrates should have higher surface free energies. However, hybridization between the magnetic *d* or *f* electron states and the substrate *d* electron states across the interface becomes a controlling factor. For instance, it has been shown that while fcc Fe(111) grows on Ru(0001) with an expanded in-plane lattice spacing that should promote ferromagnetism, the first two monolayers of Fe appear to be magnetically dead.[48] The explanation is that the in-plane expansion leads to an interplanar contraction and a strong Fe-Ru band hybridization that precludes magnetic moment formation. For the Fe/Pd(100) system, quite the opposite effect occurs. The strong *d-d* hybridization is predicted to induce ferromagnetism in the Pd substrate.[49] It is interesting to note that these trends are mirrored in the behavior of dilute Fe alloys in 4*d*-transition-metal hosts: Fe in Ru lacks a local moment, while Fe in Pd is the classic giant-moment system because of the polarization of Pd sites that extends many atomic shells away from the impurity site.

The structural, morphological, and growth-mode correlations with magnetic properties present an ongoing challenge to materials researchers working with monolayer and ultrathin magnetic-film structures. It is well documented for the Fe/Cu(100) system that growth-temperature and film-thickness variations change the magnetic spin orientations in a systematic manner.[46] A magnetic anisotropy diagram has been constructed (Fig. 2) to summarize the results. Subsequent studies[48] suggest that there is a degree of generality to the systematics observed in Fig. 2. However, the detailed structural underpinnings of the observed trends remain elusive.

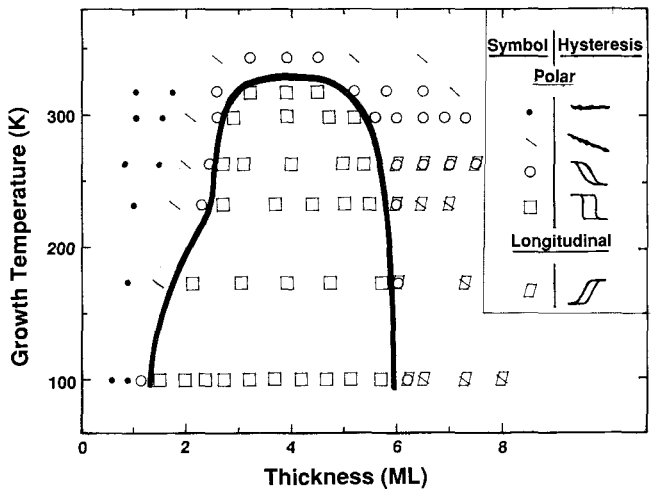

FIG. 2. The region of stability of perpendicular anisotropy for fcc Fe/Cu(100) outlined on a plot of growth temperature versus film thickness in monolayers (ML). The Kerr-effect measurements used to determine the stability boundaries were made at the growth temperature.[46]

The perfection of surface structures impacts on the study of critical phenomena in two dimensions as well. Imperfections can prevent the magnetic correlation length from diverging as the critical temperature is approached. This broadens the transition and couples the data-analysis task of defining the magnetization exponent to that of simultaneously defining an effective T_C.

Surface perfection also manifests itself in the quest to verify the theoretical predictions regarding possible ferromagnetism[50–52] at the {100} surfaces of Cr. This prediction also indicates that the moments are dramatically enhanced at the surface. The surface-ordering temperature is also raised, relative to the Néel temperature of bulk Cr. The enhanced surface magnetism of Cr(100) leads to ferromagnetic (100) sheets that are coupled antiparallel to each other on adjacent layers.[50–52] The problem is that if terrace widths at the surface are smaller than the domain-wall thickness, the surface becomes divided into antiparallel domains, and there is no net moment on a macroscopic scale. Since even a (100) surface well-defined by standard surface-science criteria does have step densities of order one per 100 Å, even with polarized-electron imaging of the domain structure the present resolution level (~500 Å) is insufficient to clarify this exciting issue. The future should bring increased experimental resolution in imaging and an enhanced ability to create ultraflat surfaces, e.g., by epitaxy on GaAs-based heterostructures, or via homoepitaxial smoothing of the surface as part of the fabrication process.

2. Metastable epitaxial films

Elemental magnetic materials exist in a variety of crystallographic and magnetic phases. Thin-film growth of these materials on crystalline substrates allows the forces present at the interface to drive the film into specific crystallographic structures. These structures may be a known high-pressure or high-temperature phase, or a phase not previously observed. Since the energies associated with a change in crystal structure (≈0.1 eV per atom) are of the same order of magnitude as energies associated with a change in magnetic structure (e.g., ferromagnetic to antiferromagnetic), often the magnetic properties of thin films dramatically depend on the growth conditions and structure of the substrates. These artificial magnetic materials, which are stabilized by their growth in thin film form, are collectively referred to as metastable structures and now form the basis of an active field of research. They greatly expand the number and variety of magnetic materials by essentially making new materials from "old" elements.

In addition to providing new structures, these metastable phases provide stringent tests of calculational techniques used to predict structural and magnetic properties of magnetic materials. These techniques (see Sec. II, Theoretical Background) are capable of yielding the total energy of an elemental crystallographic system as a function of lattice structure and spacing, including a zero-temperature prediction of magnetic moment and magnetic structure. Indeed, the failures of current calculations to predict accurately the energy hierarchies of these phases are helping to pinpoint the deficiencies in the theoretical underpinnings.

An example of the richness of phases available to a magnetic element is provided by inspecting the phase diagram[54] of Fe shown in Fig. 3. At ambient pressure and temperature the common bcc form of ferromagnetic iron is obtained. At high pressure and low temperature, however, the hcp ϵ-phase, which is nonmagnetic, is predicted. This is the expected phase in the absence of magnetic effects, as given by the structure of the other elements in the iso-electronic sequence, Ru and Os. At higher temperatures paramagnetic fcc γ–Fe and bcc δ–Fe are predicted, and at even higher temperatures the system melts.

The total energies calculated[55] for the cubic phases are shown in the top frame of Fig. 4. These calculations show a clear energy minimum for bcc Fe at the observed lattice constant and correctly predict it to be ferromagnetic. A nonmagnetic fcc phase is predicted for a smaller Wigner-Seitz radius at slightly higher energy and a second ferromagnetic fcc phase at a larger Wigner-Seitz radius at much higher energy. (These calculations were based on spherical approximations to the atomic potentials and charge densities; more accurate

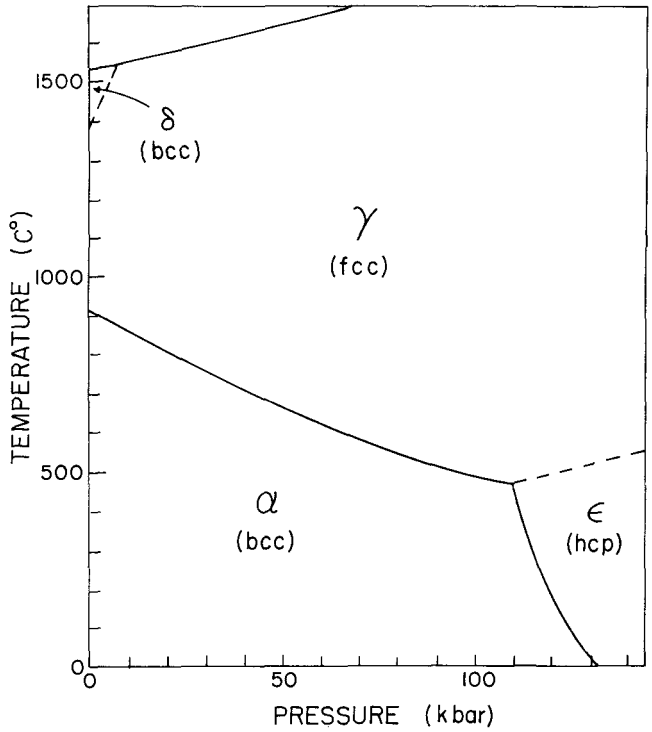

FIG. 3. Phase diagram for bulk Fe.[54]

nonspherical calculations yield, erroneously, the fcc phase as the most stable one for iron.[9]) The point at which these two branches cross corresponds to a lattice constant very close to that of fcc Cu (bottom frame of Fig. 4). Epitaxial growth of Fe on a Cu substrate has shown that either ferromagnetic or antiferromagnetic fcc Fe can be obtained, depending on the detailed conditions of growth (substrate temperature, surface preparation, and surface cleanliness).[45] This indicates that even fine details of total energy calculations may be manifest in metastable thin films.

Another example of a metastable phase is given in the second frame of Fig. 4. It shows total energy curves for two cubic phases of Co. Face-centered cubic Co is the high temperature ferromagnetic phase observed in nature; however, there is no naturally occurring bcc phase of Co. Experimentally, however, a bcc ferromagnetic phase[53] was successfully formed by epitaxial growth on GaAs. Total-energy calculations[55] yield the bcc-Co phase with the observed lattice constant, shown in Fig. 4, and correctly predict it to be ferromagnetic.

Finally, Fig. 4 indicates that there should be bcc phases of Ni, both ferromagnetic and nonmagnetic, even though in nature Ni appears only in a ferromagnetic fcc phase. Body-centered cubic Ni has been reported to be stabilized by epitaxial growth on a single crystal surface of Fe(100). At this lattice constant, it is far from the metastable equilibrium value for the Wigner-Seitz radius indicated by the calculation, and

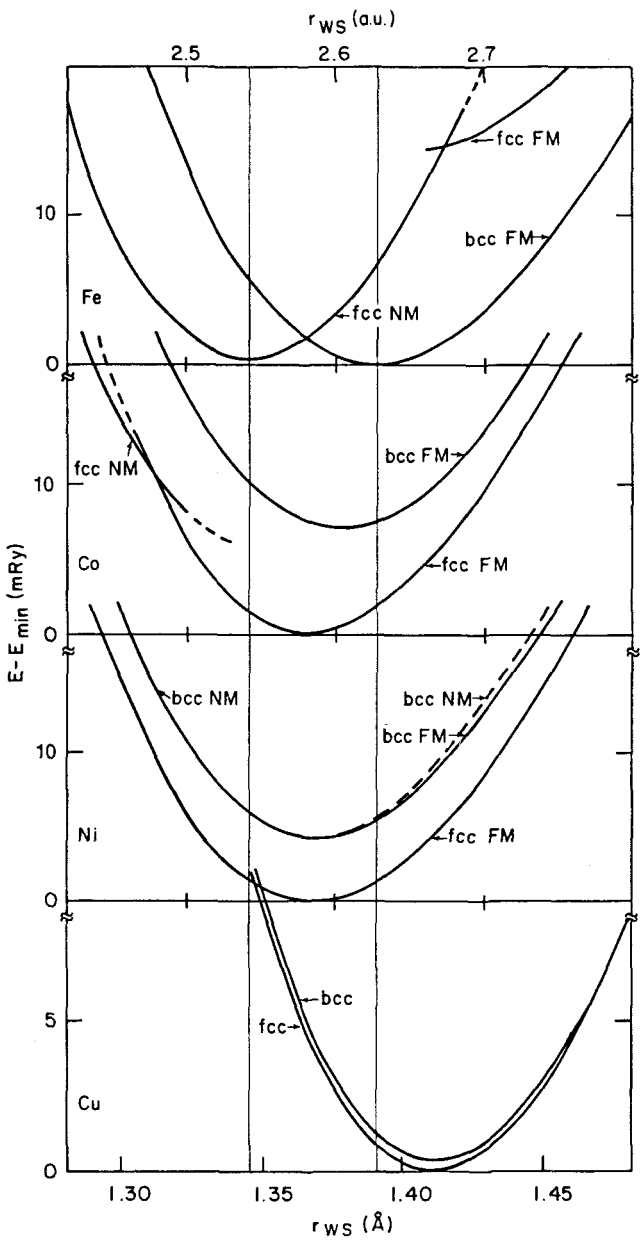

FIG. 4. Calculated total energy versus Wigner-Seitz cell radius for several metals.[55]

the strong influence of the ferromagnetic substrate made magnetic characterization difficult.

3. Semiconductor substrates

Single-crystal semiconductor substrates provide a very attractive template for the epitaxial growth of metal films. In particular, a group consisting of Ge, GaAs, AlAs, and ZnSe all have lattice constants very close to 5.65 Å. This is also very close to twice the lattice constant of bcc Co (2.82 Å), bcc Fe (2.87 Å), and bcc Ni (2.89 Å), which should permit a c (2 × 2) reconstruction of the metal films upon these substrates. Although bcc Co has been successfully grown on GaAs[53] and bcc Fe on Ge, GaAs, and ZnSe,[56] there is an important issue of interface chemistry with these systems. It has been observed, for example, that although Fe films grown on Ge have excellent structural quality, there is considerable interdiffusion at the interface, which diminishes the magnetic moment. When Fe is grown on GaAs, photoemission studies[57] show that FeAs is formed at the interface, releasing a partial monolayer of Ga which is then covered by subsequent deposition of Fe. Commercially processed substrates, however, can release significant amounts of As which largely diffuse to the top surface of the Fe film during growth. Furthermore, the small amounts of As incorporated in the film within an exponential decay length of 10 Å from the surface show an inordinately large effect in diminishing the moment up to 100 Å from the interface. These effects can be eliminated by first capping the substrate with a homoepitaxial layer of GaAs or an epitaxial layer of ZnSe. The growth of Fe on ZnSe epilayers shows the full Fe moment in films down to 20 Å thickness and the cubic anisotropy of bulk Fe.

4. Rare earths

The growth of rare earths provides a particularly fertile ground for the study of magnetic phenomena in thin films and their relationship to magnetism in reduced dimensionality. The main reason is that rare earths display a variety of systems which are chemically similar, span a large range of ionic radii and crystal structures, and present a wealth of magnetic structures including helical, ferromagnetic, antiferromagnetic, and cone magnetic structures. In addition, rare earths exhibit a great variability of thermodynamic phase diagrams ranging from complete immiscibility—as is the case for many rare earths with transition metals—to the formation of complete sets of solid solutions—as is the case of two rare earths. The epitaxial growth of rare earths and transition metals is particularly challenging because of the high reactivity of the rare earths and the high melting points of many of the transition metals. As a consequence, MBE is used for these systems, with special care taken to avoid contamination. Of course, as in all MBE growth, the structure of the epitaxial layer is monitored *in situ* using RHEED, RHEED oscillations, and *ex situ* x-ray and neutron diffraction. Generally it has been found that the growth of rare earths can be accomplished quite conveniently on a transition metal, for instance Gd on W(110),[58] Y on Nb(110),[59] or Ce on V(110).[60] One reason that these systems can be grown with relative ease is that they do not form solid solutions in their phase diagram, and possibly this facilitates the growth of a segregated rare earth.

The growth of epitaxial rare-earth films and multilayers had as a key ingredient the discovery[59] that

Y(0001) grows epitaxially on Nb(110). Following the growth of 50–100 nm of Y(0001), excellent rare-earth films and multilayers can be produced. It is currently possible to produce films and multilayers whose crystalline perfection, as measured by Bragg peak width and mosaic spread, rivals that of bulk single crystals of the same elements. The elements Gd, Dy, Er, and Ho have been extensively studied, as have multilayers of the same elements, separated by yttrium, rare-earth-yttrium alloys, or other rare earths and alloys.[39,41,61–63] A recent development in this field uses homoepitaxy to produce a chemically clean surface on a substrate material, which can have any desired crystallographic orientation. For instance, crystals of hexagonal Y are cut to expose $\{10\bar{1}0\}$ and $\{11\bar{2}0\}$ faces after which additional Y is grown epitaxially. These have then been used as substrates on which to grow $\{10\bar{1}0\}$ and $\{11\bar{2}0\}$ rare-earth films and multilayers.[64] Such samples are important for the study of the influence of epitaxial constraints on magnetic properties, and open a new field for the study of propagation of magnetic order along different crystallographic directions.[65] This technique has also been used to produce rare-earth films and superlattices on thin substrates. Niobium foils were grown as (110) single crystals (1 $cm^2 \times 5\ \mu m$),[64] followed by the same procedure used to grow rare earths on sapphire substrates. Such samples are particularly useful for x-ray scattering studies in transmission geometry, for mechanical and thermodynamics measurements, and for electron microscopy of epitaxial samples.

The epitaxy of rare earths on vanadium has been pursued in order to understand the role that lattice matching and chemistry play in the growth.[60] This system exhibits complete immiscibility with most rare earths and therefore it was possible to study how its growth is affected by lattice mismatch. In particular, Ce(111) on V(110) has shown the presence of a new epitaxial orientation, not yet observed nor predicted in any fcc(111)/bcc(110) system,[66] and the stabilization of a new metastable phase of Ce, expanded in the directions parallel and perpendicular to the growth direction. The growth of dysprosium on vanadium exhibits a variety of novel expanded phases for dysprosium as well as a series of surface reconstructions as a function of thickness.[67] It is quite interesting to note that the expanded phases are not governed by the Poisson's ratios of the overlayer, and that the expansion occurs in all directions, similarly to the earlier observations for the growth of Fe on GaAs, where a contraction occurs in all directions.

5. Oxides

One particular type of system which is of great importance and which has not been studied extensively is the growth of epitaxial oxides. Oxides in many cases exhibit interesting magnetic properties—such as antiferromagnetism—and are the basis for a variety of devices, especially when used in conjunction with a ferromagnetic material. The growth of oxides has also received enhanced notoriety because of the discovery of high-temperature superconductivity in ceramic oxides.[68] The growth has usually been accomplished using oxygen sources in an MBE system, using reactive sputtering or laser ablation techniques. Nickel monoxide (NiO) and cobalt monoxide (CoO) single crystals have been prepared on MgO substrates using CVD.[69] Recently titanium oxides were grown on sapphire by MBE using activated oxygen sources.[70] Chemical vapor deposition was used to prepare a variety of thick oxide films, especially ferromagnetic compounds such as NiO, CoO, $Ni_xCo_{1-x}O$, and $RFeO_3$ (where R is a rare earth).[71]

6. Multilayers

A large variety of multilayered systems have been grown: ferromagnetic-normal metals, ferromagnetic-superconducting, rare-earth-rare-earths, etc. The preferred growth method has been sputtering of MBE, although recently titanium-oxide-titanium superlattices have been grown by the CVD techniques described earlier.

Multilayered systems which are lattice-matched have been grown by thermal evaporation or MBE. The MBE grown, lattice-matched systems exhibit narrow x-ray diffraction lines comparable to the instrumental resolution. The lattice mismatched systems are generally textured and exhibit broader x-ray diffraction lines. However, questions regarding interfacial chemistry have not been fully addressed, because detailed understanding of roughness, disorder, and interdiffusion is only now being addressed (see Sec. IV, Techniques and Facilities).

D. Physical properties

1. Proximity and interfacial effects

In some systems, interface effects of a purely magnetic origin extend beyond the interface and into the bulk, thus giving rise to a proximity effect. Examples can be found in transition-metal systems where one side consists of a strong ferromagnet, such as Co, and the other side consists of an easily polarizable (almost magnetic) material, such as Pd, or a weakly magnetic material, such as Cr. The strong electron-electron interaction of the fully saturated ferromagnet, frustrated by a lack of *d* holes from producing a larger moment, induces through hybridization and exchange an additional magnetic moment in the *d* bands of the polarizable material. This effect is analogous to the polarization of the Fe atoms in dilute Fe–Co alloys[74] and the polarization of Pd atoms in dilute Pd–Fe alloys.

Theoretical and experimental studies of proximity effects in transition and simple metals have established a series of empirical rules that can be summarized as follows[75–77]:

(1) The magnetic moments of cobalt and nickel are virtually saturated; they can be only very slightly changed by their immediate environment. The fractional change, however, can be appreciable in nickel (which has a small moment of about 0.6 Bohr magnetons), but is negligible in cobalt.

(2) The magnetic moment of iron, which has only a moderate electron-electron interaction, can be appreciably affected by its immediate environment.

(3) Chromium, which is a weak magnetic ion, may have its moment profoundly altered by the presence of surfaces, interfaces, and both magnetic and nonmagnetic neighbors.

(4) The "almost magnetic" elements, vanadium and palladium, may acquire a sizable magnetic moment in the proper environment.

(5) Free surfaces, which reduce the local bandwidth of a metal, tend to increase the magnetic moment of an element; hence the surface of chromium has a much larger moment than the bulk,[78,79] nickel tends to be marginally more magnetic at the surface,[80] and it is possible that some crystallographic faces of vanadium exhibit a magnetic moment.[75]

(6) Proximity of a nonmagnetic metal tends to suppress the magnetic moment of some elements; this effect depends crucially on the overlap of the wave functions between the d band of the magnetic metal and the conduction band of the nonmagnetic one.

(7) The proximity of a strongly magnetic element tends to induce or enhance magnetic moments on the neighboring, susceptible elements. Thus iron becomes more magnetic in the proximity of cobalt,[74,81] the enhanced moment of the chromium surface tends to propagate over several layers into the bulk,[82] chromium acquires a large moment in the proximity of iron[78] and/or cobalt, and vanadium and palladium may develop sizable magnetic moments in the proximity of iron and/or cobalt.

2. Exchange coupling across interfaces

Magnetic exchange coupling between ferromagnetic and antiferromagnetic layers was originally discovered in oxidized Co particles.[83] The antiferromagnetic CoO surface layer is exchange-coupled to the ferromagnetic Co interior which results in an imposed unidirectional anisotropy. As a result, an asymmetric hysteresis loop shifted from zero field by the exchange bias field H_b developed. A second thin-film system which has been extensively studied[84] is ferromagnetic permalloy, $Ni_{81}Fe_{19}$, coupled to the antiferromagnetic alloy, FeMn. For a ferromagnetic/antiferromagnetic coupled system to exhibit a macroscopic exchange bias field H_b, the Néel temperature of the antiferromagnet must be lower than the Curie temperature of the ferromagnet.[85] In addition, the magnetic anisotropy energy of the antiferromagnet must be large compared to the interfacial exchange coupling so that the antiferromagnetic spin system remains substantially blocked when the magnetization of the ferromagnet rotates upon application of an external field.

The magnitude of the interfacial exchange coupling energy, E_a, is much larger in the Co/CoO system than in $Ni_{81}Fe_{19}/Fe_xMn_{1-x}$. In the latter case the magnitude of E_a is approximately 100 times smaller than expected in the simplest model: an antiferromagnetic structure comprised of uncompensated ferromagnetic layers whose magnetization, directed normal to the interfaces, alternates in sign from layer to layer.[86] Assuming no relaxation of the antiferromagnetic structure at the interface, H_b is given by (J/M), where J is the interfacial exchange coupling and M is the magnetization of the ferromagnet. Values of J comparable to those in the ferromagnet or the antiferromagnet yield, for H_b, values 100 to 1000 times greater than those observed experimentally. The experimental data are surprisingly consistent for several $Ni_{81}Fe_{19}/Fe_xMn_{1-x}$ films prepared by a number of different workers under quite different conditions.

Various models have been proposed to account for the large discrepancy between experiment and theory noted above. However, none of them can explain all the properties of this coupled system. It is quite clear that no domain wall is formed in the ferromagnetic layer via some sort of "wetting" to the antiferromagnetic layer upon rotation of its magnetization. The possibility of a planar domain wall in the antiferromagnetic layer can be ruled out, since antiferromagnetic layer thicknesses thinner than a typical domain wall by an order of magnitude give the same H_b. The most complete model proposed so far[86] suggests that interfacial atomic roughness would reduce the exchange coupling energy. Therefore roughness or chemical inhomogeneities at the interface are assumed to give rise to random interfacial exchange interactions (parallel or antiparallel to the direction of the unidirectional anisotropy). The antiferromagnet minimizes its energy by breaking up into lateral domains whose size is approximately that of the antiferromagnetic domain-wall width. Thus, averaging over the random exchange fields in a single domain implies that the interfacial exchange coupling energy is reduced by $N^{1/2}$, where N is the number of atoms in the interface layer of a single antiferromagnetic domain. This model thus predicts that the larger the antiferromagnetic domains, the greater the extent to which the random fields cancel one another. The observed values of H_b in $Ni_{81}Fe_{19}/Mn_xFe_{1-x}$ are reasonably consistent

with the model. Further refinements, including a more detailed description of the antiferromagnetic structure of MnFe, are an important prerequisite for improved understanding of this exchanged coupled system. The details of the interfacial structure and their effect on magnetism are a crucial part of this understanding.

Not only do ferromagnetic/antiferromagnetic coupled systems display a fascinating range of properties, but the interfacial exchange coupling can be harnessed to study the properties of the antiferromagnetic layer. It is extremely difficult to measure many fundamental magnetic properties of ultra-thin antiferromagnetic films, including, for example, their Néel temperatures, because of the difficulty of coupling to the sublattice magnetization. Most electron, optical and neutron scattering, and magnetic resonance techniques are incapable of examining antiferromagnetic thin films. Spin-polarized photoelectron diffraction is one of the few techniques with some potential for such studies. This technique, however, is in its infancy and is sensitive only to the magnetic short-range order which, for the two antiferromagnetic single-crystal systems so far studied,[87] persists to temperatures several times larger than the bulk Néel temperature.

The ferromagnetic layer in a ferromagnet/antiferromagnet couple forms a natural probe of the antiferromagnetic system. By monitoring the temperature at which the exchange bias field goes to zero, the blocking temperature of the antiferromagnet can be determined. This temperature is slightly lower and closely related to the Néel temperature T_N of the antiferromagnet. At a temperature just below T_N the anisotropy of the antiferromagnetic layer becomes too weak compared to the exchange coupling energy to maintain the rigidity of the antiferromagnetic lattice, which thus becomes free to follow the magnetization of the ferromagnetic layer. The dependence of the blocking temperature on the thickness of the FeMn layer in polycrystalline films of $Ni_{81}Fe_{19}/Fe_xMn_{1-x}$ has been determined[88] for a constant NiFe thickness of about 60 Å. The blocking temperature is independent of thickness for FeMn thicknesses greater than about 100 Å, but it is lower for thinner layers. The thickness dependence has been found to follow a simple finite-size scaling relationship. This method can clearly be applied to other antiferromagnetic systems and should prove to be a rich area for further work.

In contrast to the ferromagnetic/antiferromagnetic coupled systems, the magnitude of the exchange coupling in ferromagnetic/ferromagnetic systems can be very large. A wide variety of systems has been studied; they include, however, very few studies on well characterized single crystals. One example of the latter are single crystals of bcc Ni/Fe bilayers.[89] For Ni layer thicknesses greater than six monolayers the Ni lattice reconstructs and, via exchange coupling to the Fe layer, imposes a large in-plane fourfold anisotropy on the Fe layer. Less well characterized systems include polycrystalline $Ni_{81}Fe_{19}/Fe$ superlattices and a wide variety of amorphous rare earth–transition metal (RE/TM) alloy films coupled to other RE/TM alloys or polycrystalline films of Fe, Co, or $Ni_{81}Fe_{19}$. The latter systems all have been developed for their possible application in a variety of magnetic recording devices. Superlattices of $Ni_{81}Fe_{19}/Fe$ with layer thicknesses in the range, for $Ni_{81}Fe_{19}$ and Fe respectively, of ≈100–500 Å and ≈300–1500 Å combine the high saturation magnetization with the high permeabilities required for magnetic recording-head applications.[90] Whereas in these superlattices the saturation magnetization is simply the appropriate averaged saturation of the $Ni_{81}Fe_{19}$ and Fe layers, the coercivity of the superlattice is much closer to that of $Ni_{81}Fe_{19}$ than to that of Fe.

There is a large body of work on exchanged-coupled RE/TM systems. Spin-polarized photoemission studies on Fe/TbFe have shown that thin Fe layers take up the perpendicular anisotropy of the amorphous TbFe underlayer, with hysteresis loops which reflect strong exchange coupling between layers.[91] Exchange coupling between $Ni_{81}Fe_{19}$ and TbCo leads to exchange-shifted hysteresis loops with bias fields[92] as large as 500 Oe for $Ni_{81}Fe_{19}$ layer thicknesses of about 400 Å. A variety of schemes for taking advantage of the magnetic exchange coupling between two different RE/TM alloys has been proposed[93]; these schemes optimize the performance of a magneto-optic storage medium. In particular, one of the RE/TM layers is chosen to have a high magneto-optic rotation, θ_K, in the wavelength range of interest whereas the second layer, which may have a small θ_K, is chosen to have a high coercivity. Thus the first layer has optimum read-out properties and the second, optimum storage properties.

3. Coupling through nonmagnetic layers

One of the most interesting ferromagnetic/metal/ferromagnetic systems is Fe/Cr/Fe, where it has been found that successive Fe layers, for thin Cr layer thicknesses, are coupled antiparallel to each other. The coupling diminishes as the Cr layer increases in thickness. The coupling mechanism is, at present, not yet well understood. It is apparently too large a coupling to be accounted for by magnetostatic effects. Such an antiparallel coupling was observed in Fe/Cr/Fe trilayer structures[94] in spin-polarized electron-scattering studies and in Magneto-optic-Kerr-effect (MOKE) and Brillouin scattering studies.[95]

Recently the same phenomenon has been observed in MBE grown Fe/Cr superlattices by magnetization studies. The superlattice has no moment in zero field,

but the moments of the antiparallel, neighboring Fe layers can be aligned by application of fields of up to 20 kOe for Fe layer thicknesses of 30 Å, which implies very large effective exchange-coupling energies. An important property of these structures is the large drop in resistance observed on aligning the Fe layer moments.[96] This "giant" magnetoresistance effect is not yet fully understood; it is of great interest for potential recording head applications.

There have been numerous studies of systems of the ferromagnet/metal/ferromagnet type, ranging from attempts to vary the coercivity of ferromagnetic films by lamination for magnetic recording applications, to studies of single-crystal superlattices,[97,98] such as Fe/Ag. Exchange coupling of successive Fe layers in this system has been inferred[98] from the temperature dependence of the magnetization at low temperatures. A calculation[99,100] of the temperature dependence of the magnetization in the spin-wave regime for an arbitrary multilayered magnetic structure has shown that there always exists a range of temperature for which the magnetization varies as $\alpha T^{3/2}$, where the coefficient α depends on the exchange coupling between the magnetic layers. The method of calculation can be applied to obtain the exchange coupling in ferromagnetic/metal/ferromagnetic systems. In some recent elegant experiments[101] the coupling between a surface layer of $Ni_{81}Fe_{19}$ and an underlying thick $Ni_{81}Fe_{19}$ layer (separated from each other by submonolayers of Ta) was obtained.

Tunneling between a spin-polarized superconducting film coupled to a ferromagnetic layer has been extensively used to study the magnetic properties of thin ferromagnetic layers.[102] It has been proposed[103] that tunneling between two ferromagnets could depend on the relative alignment of the magnetization of the two ferromagnetic layers; this effect was subsequently observed[104] in the system Fe–Ge–Co. The magnitude of this magnetic tunneling-valve effect was found to be about half that expected from the spin polarizations in Fe and Co as deduced from tunneling[105] in ferromagnet/insulator/superconductor junctions. More recently, similar effects have been observed[106] in Ni/NiO/Co tunnel junctions.

4. Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling

Bulk rare-earth elements and their alloys with yttrium exhibit complex spin arrangements caused by the combination of strong crystal field effects and the oscillatory exchange interaction modulated by the conduction electrons (RKKY interaction). Early work in rare-earth multilayers[107,108] demonstrated that RKKY polarization propagates across the rare-earth/yttrium (0001) interface, and thus it decays slowly enough to provide coherent exchange coupling across as much as 130 Å of Y. Spiral (transverse) and c-axis (longitudinal) polarizations are preserved. Experimental results of magnetic neutron scattering, which provide evidence for the propagation of magnetic order through the nonmagnetic Y, are shown in Fig. 5. Intriguingly, the periodicity of the spin polarization in the Y is that of dilute rare-earth–yttrium alloys, while that in the rare earth deviates from bulk values at low temperatures.

A model for RKKY coupling has been proposed.[109] Rare-earth sheets were required to be immersed in the Y conduction band, but to interact with the *s-f* interactions appropriate to the rare earth. The polarization, therefore, is formed by the nesting features of the Y band structure. In the case of spiral structures, two transverse polarization waves, out of phase by one lattice spacing, are produced, thus providing a helical arrangement. A later refinement of this picture postulates the existence of a superlattice band, with the oscillating polarization being a feature of the hybrid band. Wave vector conservation parallel to the interface prevents mixing of states at the Fermi surface with different interface momenta, and can result in localization of certain electron states on one side of the interface.

Recent experiments have explored the RKKY coupling across $(10\bar{1}0)$ and $11\bar{2}0)$ interfaces. For Dy/Y the polarization is insufficient to bring the spiral order of successive rare-earth blocks into coherence but does provide adequate coupling to produce long-range ferromagnetic order in Gd superlattices. This may simply reflect the strongly anisotropic range of RKKY oscillations[64] in Y but may also be evidence for total reflection of those conduction electrons most important in providing the RKKY coupling, as presented by the superlattice band approach of Ref. 109. Other evidence for spin-dependent transmission has been seen in magnetotransport experiments in the Fe–Cr–Fe system.

5. Magnetoelasticity

The presence of strain has been used to modify the physical properties through the magnetoelastic effect. This is particularly important for materials such as rare earths and Laves-phase alloys, where magnetoelastic effects are large. This effect was first discovered in Dy superlattices[39] and films,[67] where the magnetoelastically driven ferromagnetic transition is suppressed. Similar effects have been observed in Er films and superlattices. Both Dy and Er epitaxial materials can be driven to ferromagnetism at a critical value of the applied field that depends on film thickness. In the case of Er, a variety of commensurable spin states are induced at low temperatures by fields below the critical value.[110] Bulk behavior is not recovered in films up to 1 μm thick. The treatment of this problem to date has relied on bulk values of the magnetoelastic coupling constants subject to rigid clamping assumptions. Mea-

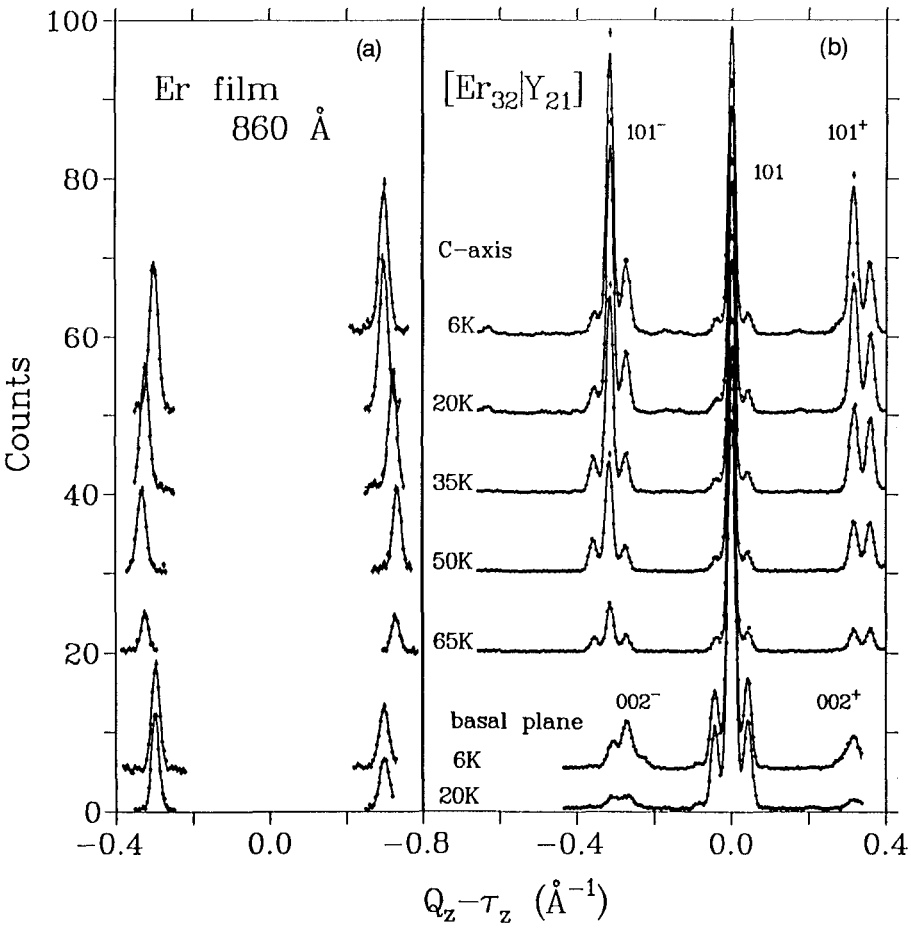

FIG. 5. Magnetic neutron scattering from (a) an Er film, and (b) an Er/Y multilayer with 32 atomic planes of Er and 21 of Y. The multiple peak structures near ±0.3 $Å^{-1}$ demonstrate the coherent propagation of magnetic order through the nonmagnetic yttrium interlayers.[110]

surements of actual strains in epilayers, along with a proper treatment of magnetoelasticity for such systems, are clearly required.

A direct measurement of the RKKY coupling can be made by studying the break-up of long-range coherence by applied fields at temperatures where magnetoelastic effects are most important. Because there is a net moment in incommensurable spiral layers, applied fields align these moments, directly acting as a random field on the spiral. The loss of coherence in Dy/Y typically occurs[39] on application of fields of the order of 5000 Oe.

6. Superlattice effects

Many of the effects described above can be conveniently studied in multilayered films since they consist of a superposition of single films.[111] Moreover, multilayers provide the possibility of *ex situ* studies without concern regarding contamination, since they can be grown very thick (~1 μm) compared to usual contamination depths. It should also be pointed out here that multilayers offer an additional dimension in characterization, as described in Sec. IV (Techniques and Facilities). The drawback is, of course, that by its very nature any single, bi-, or tri-layered film effect can be obtained only in a statistical sense, averaged over many (hopefully equivalent) repetitions of the system.

There is, however, a class of effects which cannot, even in principle, be observed in a small number of layers because they rely on the periodic nature of the multilayer. These are the so-called superlattice effects. The original observation of this type of effect was that of phonon folding in semiconductor superlattices.[112] In metal systems there have been several unobserved theoretical predictions of minigaps in the electrical transport phenomena,[113] of localized states in these gaps, and of gaps in the continuum of the density of states in superconductors.[114] These effects all rely on the presence of extended electronic states in the growth direction. However, all metal systems studied to date exhibit large amounts of interfacial scattering, as indicated by the thickness-dependent resistivity.[42] Whether this scattering is sufficient to break down the existence of extended states perpendicular to the layers and in effect confine the electrons to individual layers is not clear at this time. Possibly these effects should be observable in high-perfection superlattice systems which exhibit no interfacial scattering.

A superlattice effect which does not require perfection at the atomic level is the development of the

magnon bands in ferromagnet/normal-metal superlattices.[115] The coupling in these types of superlattices depends on the long-range dipolar interaction which is not much affected by small amounts of disorder at an interface. The individual modes in each one of the magnetic layers spreads into bands of magnons as the intervening normal metal thickness is decreased. This is illustrated in Fig. 6 where a qualitative plot of magnon frequency versus normal-metal separation is shown. These predictions are conceptually similar to the development of energy bands in a metal from the discrete electronic levels present in individual atoms. Detailed theoretical predictions[115] have been obtained, including the dependence of the magnon frequencies on layer thicknesses, magnetic field, saturation magnetization, and wave vector. All these have been verified in detail in a series of measurements in Ni/Mo superlattices.[116] As an example, Fig. 7 displays the magnon frequency as a function of magnetic field for a number of superlattices. The excellent agreement between experiment (plus signs) and theory (solid line) shows that our understanding of this phenomenon is on a firm footing. It is even more striking that all parameters (thicknesses, magnetic field, magnetization, wave vector) that enter into the calculations are independently measured so no parameters need to be adjusted to bring theory and experiment into agreement.

IV. TECHNIQUES AND FACILITIES

The new scientific opportunities in magnetic-materials research are ripe for exploration because of the modern research techniques and facilities available

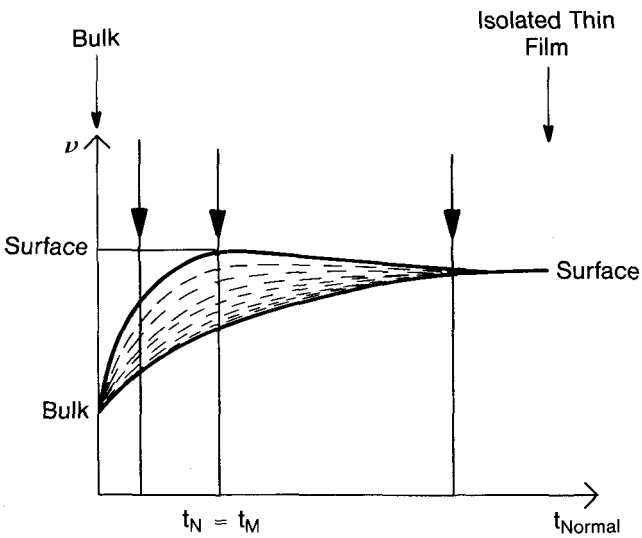

FIG. 6. Qualitative development of magnon bands in a magnetic/normal-metal superlattice. The figure shows the magnon frequency as a function of normal-metal thickness t_N; t_M is the magnetic-metal thickness. The shaded area between the two lines represents the band of superlattice modes.[115]

to the materials science community. The techniques span the range from the structural-characterization tools shared with the semiconductor heteroepitaxy fabrication and processing community, such as Reflection High-Energy Electron Diffraction (RHEED), to the inherently surface-sensitive probes of magnetism, such as spin-polarized electron spectroscopies, to traditional probes of bulk magnetic materials that are adapted to enhance their surface sensitivity, such as Mössbauer spectroscopy by means of conversion-electron detection. The techniques may make use of major facilities with accelerator-based photon sources, neutron scattering facilities, and high-energy electron microscopes. The structural techniques shared with the semiconductor community have been covered in an independent panel report.[117] The present contribution describes methods used, almost exclusively, by the magnetic-materials community.

While there is a considerable number of sophisticated instruments devoted to the analysis of ultrathin magnetic films, most individual university laboratories do not have the resources to obtain or support all, or even most of them. In addition, many of these instruments do not measure magnetic properties directly, at high frequencies, or at very rapid rates. And yet many of the applications, at least for soft or semi-soft magnetic materials, involve high-speed switching. Similarly, scientists need data which are straightforward to interpret and a rapid turnaround during materials studies. The recent developments of the alternating gradient[118] magnetometer have demonstrated that simple, inexpensive techniques can be developed which have the sensitivity to extend the range of measurements to very small samples. Extension of this method to even smaller samples and/or to high-frequency regimes would enable many of the smaller laboratories to contribute to the understanding of surface magnetism. In the same fashion, since surface anisotropy is believed to play such an important role at the interface, increased sensitivity improvements in torque magnetometry, and the ability to use instruments *in situ* need to be developed. Another excellent example of the usefulness of a recently developed simple instrument, which can be used *in situ* as the sample is prepared, is the surface magneto-optic Kerr effect technique.[119] Work to develop techniques and instruments as simple as these should be encouraged.

A. Magnetometry–spectroscopy

1. Mössbauer spectroscopy

Mössbauer spectroscopy can be used as either an *in situ* or *ex situ* technique to measure the magnetic hyperfine spectra of the magnetic atoms, particularly ^{57}Fe. The most useful approach for films is conversion

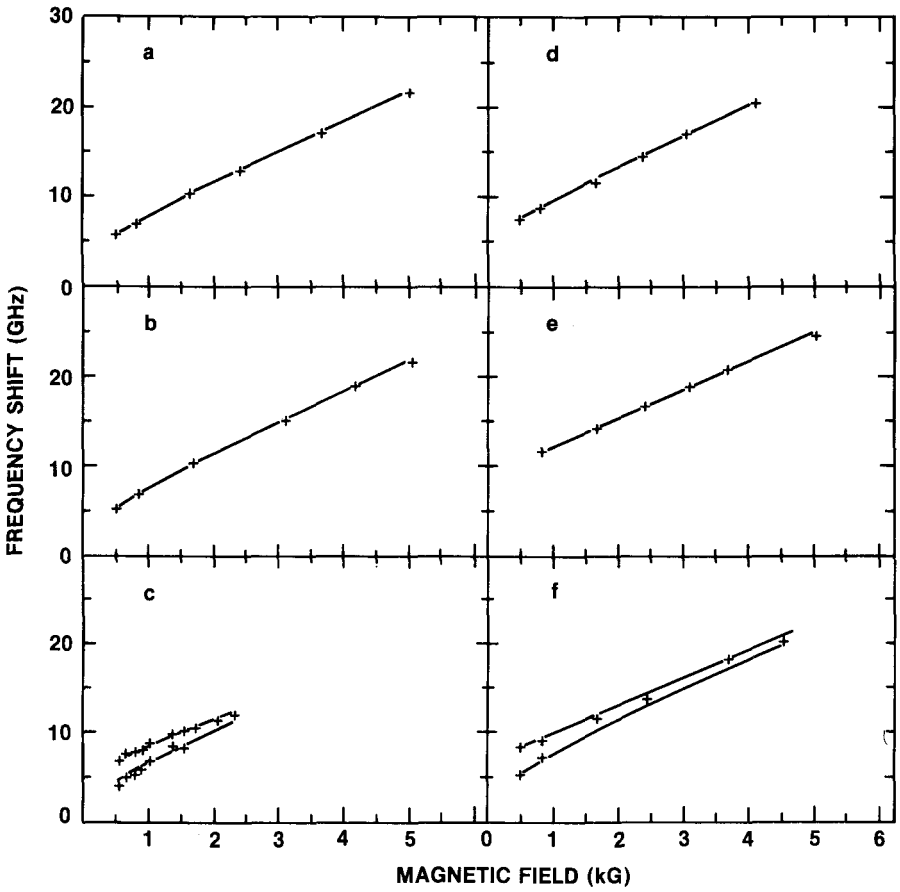

FIG. 7. Field dependence of magnon frequencies (crosses) in a representative set of Ni/Mo superlattices, together with fits to theoretical expressions (solid lines), show the existence of a true superlattice effect. The superlattices are 1–2 μm thick built of bilayers of magnetic-nonmagnetic metals with thicknesses of (a) 100 Å/300 Å; (b) 100 Å/100 Å; (c) 138 Å/46 Å; (d) 250 Å/750 Å; (e) 5000 Å/5000 Å; and (f) 540 Å/180 Å, respectively.[116]

electron spectroscopy, which is carried out *in vacuo*. One captures the photoemitted electrons and passes them through an energy analyzer to a detector. The spectra thus obtained contain not only information about the hyperfine field (which is related to the magnetic moment) but also the quadrupole splitting (indicative of deviation from cubic symmetry) and isomer shifts (a measure of the conduction electron-spin density at the nuclei). Although Mössbauer spectroscopy does not directly measure the magnetic moment, the latter can be indirectly obtained by means of detailed electronic structure calculations.

2. Magnetic resonance

Magnetic anisotropies can be very precisely and rapidly determined by means of angle-resolved magnetic resonance. Typically carried out over a range of frequencies from ≈10 GHz to 35 GHz and applied magnetic fields up to 30 kOe, not only the bulk anisotropies but in-plane and out-of-plane uniaxial surface anisotropy energies can be obtained. Furthermore, the temperature dependence of these anisotropies may be readily measured. This is important, since generally the magnetic anisotropy energy is much more temperature dependent than the magnetization itself. Ferromagnetic resonance signals have been obtained[120] from a submonolayer film of Fe grown on Ag(100) and a monolayer[121] of Gd on W. Since what is measured is the integrated absorbed power within the resonance line, anything which broadens the linewidth ultimately reduces the precision of the technique. Magnetic resonance is therefore also a sensitive measure of the quality of the film as it manifests itself in the magnetic properties. This includes uniformity of the magnetization, uniformity of thickness, presence of spin-wave scattering sites (cracks, pinholes, imperfections), and uniformity of strains.

3. Magnetometry

There have been isolated efforts to carry out torsion magnetometry and ferromagnetic resonance *in situ*; however, these techniques have not become widespread. Magnetometry and resonance are largely regarded as the primary *ex situ* characterization techniques to obtain important information about the mag-

netization and the anisotropy. A common commercial vibrating sample magnetometer (VSM) has sufficient sensitivity to measure a 20 Å, 1 cm^2 area Fe film with its full moment of 2.2 μ_B. Such instruments typically offer a sensitivity of up to 10^{-5} emu. Three orders of magnitude in sensitivity can be gained by using a commercial Superconducting Quantum-Interference Device (SQUID) susceptometer; however, at such low levels the diamagnetic signal from the substrate in general dominates the data. Very careful procedures must be employed to remove this diamagnetic signal in order to obtain information which truly represents the film.

More recent developments in magnetometry include a vibrating reed magnetometer[122] and an alternating-gradient magnetometer.[118] The former takes point-by-point measurements approaching the sensitivity of a SQUID but at lower cost. The latter also approaches the sensitivity of a SQUID but provides continuous data similar to a VSM. In addition, a simple torsion magnetometer, based on a glass fiber, can be readily constructed to yield a sensitivity sufficient to measure 1 monolayer of Fe on a 1 cm^2 surface.

4. Magneto-optics

The magneto-optical Kerr and Faraday effects date back to the latter half of the nineteenth century. Nevertheless, they are now enjoying a renaissance because of recent developments in both the basic and applications-oriented communities. On the basic side, it has recently been demonstrated that the Kerr effect can be used to detect monolayer and even submonolayer magnetism.[119] The applications-driven opportunities are associated with the commercial potential of materials for high-density magneto-optical data storage.[123] In addition, the recent development of Kerr microscopy to image magnetic domains and to observe magnetic-switching phenomena has helped revitalize the classic field of micromagnetics.[124,125]

The Surface Magneto-Optic Kerr Effect (SMOKE) provides a valuable, *in situ* characterization probe of the magnetic and magneto-optic properties of magnetic films during the growth process. The Kerr effect involves the rotation of the polarization of light reflected from a magnetized surface. The magneto-optic coupling is caused by the spin-orbit interaction. The technique requires the application of an external magnetic field to reverse the magnetization direction of the sample in the growth chamber. Otherwise, the optical components are outside the vacuum system. Typically the system consists of a laser source, a polarizing analyzer, and a photodiode detector. Magnetic hysteresis curves are obtained by monitoring the light intensity at the detector as the field is swept. To address key issues associated with the surface magnetic anisotropy, the field can be in the film plane (longitudinal Kerr effect) or perpendicular to it (polar Kerr effect). The temperature dependence of the hysteresis loops can be used to monitor the magnetization and coercivity. Quite recently the Kerr effect was used to obtain the magnetization exponent β in the critical regime for the system Fe/Pd(100), and good agreement was found with that expected theoretically for a 2-D Ising system.[126] The Kerr effect can be used as well to monitor the Curie temperature as a function of thickness, which provides a fundamental characterization parameter of the films of interest.

In the future it should be possible to use tunable photon sources in the optical-frequency region to monitor the Kerr rotation of magnetic monolayer and ultrathin, metastable phases. This form of Kerr spectroscopy will provide electronic structural information in the form of a joint density of states weighted by magneto-optic matrix elements.[127] The spectral information should complement that obtained from k-dependent probes of the band structure, such as angle-resolved, spin-polarized photoemission.

The Kerr effect is not an inherently surface-sensitive probe. The optical penetration depth in metals is ~100–200 Å. The surface sensitivity is derived from the sample fabrication techniques that create extremely thin epitaxial magnetic films. It is of interest to use complementary techniques with different probing depths to understand coupled magnetic layers, for instance. It should be possible to develop the Kerr effect into such a probe by using nonlinear optical processes; surface sensitivity will be obtained by monitoring the Kerr rotation in the Second-Harmonic Generation (SHG) mode.[128] The SHG technique has recently gained prominence as an advanced surface-analysis technique.[129]

Brillouin light scattering has also proven valuable to obtain the magnetization, and exchange and anisotropy constants from magnon spectra. These studies can be performed *in situ* on overlayers,[130] or as a post-growth characterization tool on superlattice and sandwich structures[131,132] in air or in controlled high- or low-temperature environments. The information obtained is quantitative and cross-correlates with Ferromagnetic Resonance (FMR) data.[133]

B. Polarized electron techniques

1. Spin-polarized photoemission spectroscopy

The most direct information on the ferromagnetic electronic structure at surfaces can be gained by spin-polarized photoemission spectroscopy. Early spin-polarized photoemission studies[134] measured the polarization of the photo yield as a function of photon energy without energy analysis. Such measurements still have the advantage that they can be performed as a function of applied magnetic field perpendicular to the surface

up to magnetic saturation of the sample. The intensity of synchrotron radiation permits energy analysis of the electrons photoemitted from a material magnetized in the plane of the surface (such as to minimize stray magnetic fields).[135] A movable spin and energy analyzer allows investigation along different directions in k-space. Thus, utilizing the intensity and tunability of synchrotron radiation for spin, energy, and angle-resolved photoemission, one can obtain a complete mapping of the spin-dependent band structure over the entire Brillouin zone.[136] With highly focused photon beams from undulators, it will become possible to combine spin-polarized photoelectron spectroscopy with microscopy to obtain spin-dependent electronic structure information with high spatial resolution.

With the increasing availability of high-intensity Vacuum UltraViolet (VUV)/soft x-ray radiation based on insertion devices, spin-polarized photoemission spectroscopy will play an increasing role in magnetic materials research. Studies of surface shifts in shallow core levels, e.g., $4f$ levels in rare earths, allow one to distinguish a magnetization at the surface different from underlying layers.[137] At x-ray Photoemission Spectroscopy (XPS) energies, the polarization of electrons emitted from multiplet split core levels, such as a $3s$ or $3p$ level in Fe, gives element-specific magnetic information.[138] In this sense it would be similar to polarized Auger spectroscopy, but possibly easier to interpret. Furthermore, it may be possible to extract quantitative values for atomic magnetic moments at surfaces from the spin-polarized XPS measurements.[139]

2. Polarized Auger spectroscopy

The strength of Auger electron spectroscopy as a surface analysis technique derives both from its surface sensitivity and the fact that Auger electron energies are element specific. In the case of a ferromagnet, the Auger electrons may also be spin polarized. The spin polarization results from the different occupation of the spin-split valence-conduction electrons; when these electrons at the top of the Fermi distribution are directly involved in the Auger emission process the emitted electrons are naturally polarized. If, on the other hand, only core levels are involved, there may still be a spin polarization because of the exchange interaction of the valence-electron spin density with the filled core levels. Through spin-polarized Auger, one has an element-specific probe of the local magnetization at a given site. Spin-polarized Auger spectroscopy is useful not only for investigating the magnetic properties of a surface, but it can also provide information (in films of a few layers) on the magnetic properties of substrate layers near the interface.

Some features of spin-polarized Auger spectroscopy are illustrated in the investigation[140] of the magnetic coupling of a monolayer of Gd evaporated on an Fe(100) crystal surface. The spin polarization of the Fe and Gd Auger lines shown in Fig. 8 have opposite

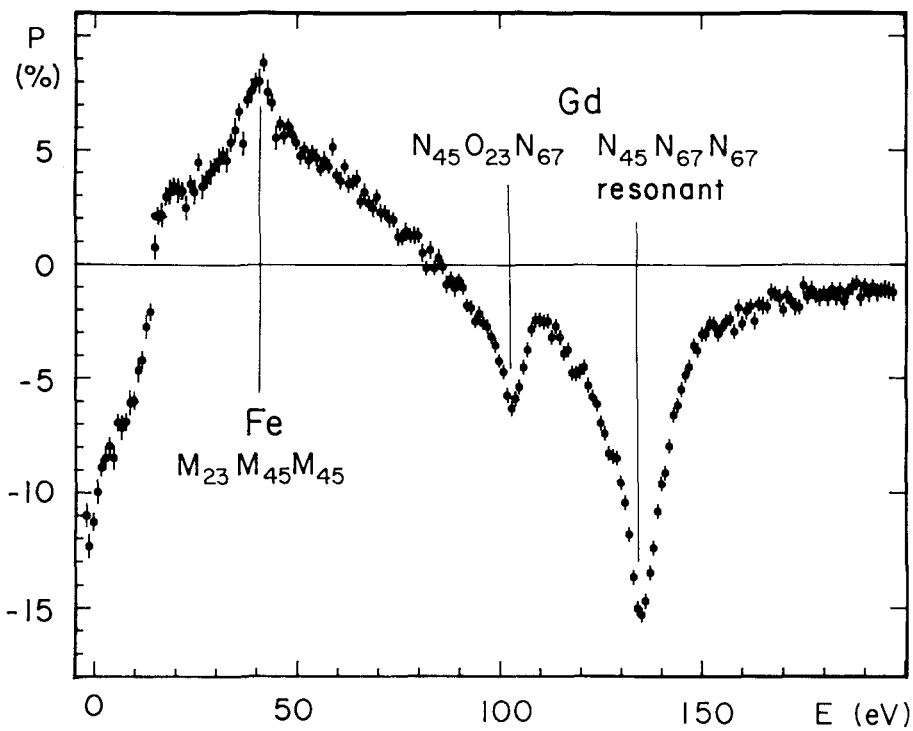

FIG. 8. Spin polarization versus kinetic energy of secondary electrons, including the labeled Auger transitions, from a Gd film on Fe(100), excited with primary electrons of 2500 eV. The film thickness is ≈1 monolayer (2.4 Å); T = 150 K.[140]

sign, indicating that the magnetic moments in the Gd overlayer are coupled antiparallel to those in the Fe substrate. In the same investigation it was possible to measure independently the temperature dependence of the magnetization of the Gd layer and the Fe interface layers, taking advantage of the Auger elemental specificity. For electron kinetic energies below 20 eV, i.e., in the secondary-electron range, the electron polarization is seen to be negative. This is surprising since low-energy secondary electrons from Fe have a positive spin polarization and is perhaps indicative of the spin-dependent inelastic scattering, anomalously large in this case, discussed in the section on polarized secondary electron emission.

3. Spin-polarized low-energy electron diffraction (SPLEED)

Low-energy-electron diffraction (LEED) is one of the standard techniques to study the structure of surfaces. Surface reconstructions and relaxations have been studied in great detail for clean single-crystal surfaces, and the geometry of adsorbates has been established for many systems.[141] Using a spin-polarized electron beam (SPLEED) on a ferromagnetic surface, one can also gain information on the surface magnetization through the additional exchange interaction potential.[142] For instance, the temperature dependence of surface magnetization in the magnon regime has been studied on a surface of a metallic glass,[143] and the critical exponent on Ni single-crystal surfaces has been determined.[144] More recently, the critical behavior of thin epitaxial Fe layers has been measured.[145] SPLEED has also been used to measure the surface magnetic moments on Fe and Ni surfaces.[146] SPLEED studies on Gd surfaces showed an enhanced surface Curie temperature T_{Cs} and indicated an antiferromagnetic surface coupling.[137]

Since the strong interplay between structure and magnetism is well known, it would be highly desirable to combine a structural tool with a probe of the magnetization. SPLEED contains information on the structure and magnetization simultaneously. Quantitative LEED structural analysis requires the comparison of intensity versus energy spectra on a number of diffracted beams with the results of multiple scattering calculations. Experimentally, large amounts of data can be accumulated by using Video-LEED systems. It would be possible to convert such a system into a Video SPLEED by adding a spin-polarized (e.g., GaAs) electron gun.[147] This would allow the detailed structural analysis and magnetic structure determination of a number of interesting systems. For example, one would be able to study the layer-dependent magnetizations at single-crystal surfaces in great detail (also their temperature dependence). The structure and magnetic moments of monoatomic layers can be studied. Also, SPLEED can give information on the magnetic moment distribution in epitaxial ultrathin films, e.g., distinguishing moments at the interface, surface moments, and center-atom moments, putting state-of-the-art electronic structure calculations to a test.

4. Spin-polarized secondary electron emission (SPSEE)

When bombarding a surface with high energy electrons (greater than a few hundred eV), a large number of low-energy secondary electrons is emitted. This low-energy secondary-electron cascade is produced by multiple inelastic scattering. If the sample has a net magnetic moment, the secondary electrons are spin polarized. It is well established that the direction of the spin polarization is aligned with (and opposite to) the direction of the magnetization. Therefore, secondary electrons can be used to determine the magnetization distribution at a surface.

The expected polarization of the low energy "true" secondary electrons, to the extent they are a representative sample of the valence electrons, is estimated to be $P = n_B/n$, where n_B is the magnetic moment per atom (Bohr magneton number) and n is the number of valence electrons. The polarization is expected to be 28%, 19%, and 5% for Fe, Co, and Ni, respectively.

There are two unexpected features in SPSEE: (1) The spin polarizations of the very low energy electrons ($\lesssim$10 eV) are enhanced by a factor of two or three compared to the average valence band polarization. This has been established for all three ferromagnetic 3d-transition metals.[148] (2) The surface sensitivity is apparently much greater than expected from the "universal" electron mean-free path curve. The magnetic probing depth in Ni and Fe is found[149] to be only of the order of 5 Å, which makes SPSEE an attractive technique for the study of ultrathin film systems. It has been suggested that both effects have their common origin in strongly spin-dependent inelastic scattering.[148] There is a need for a complete theory that would allow one to calculate inelastic scattering, with inclusion of the exchange interaction.

By using highly focused primary beams (electron microscopes), the magnetization at a surface can be mapped with high lateral resolution (100 Å) (see the subsection below on SEMPA). Even without the spatial resolution provided by the Scanning Electron Microscope (SEM), the measurement of the polarization of secondary electrons provides a strong signal and a good way to obtain information on the average magnetization at a surface, as opposed to the small sampling of particular transitions over a small region of k space as observed in polarized photoemission, and without the complications introduced by diffraction or multiple

scattering as in Spin Polarized Low Energy Electron Diffraction (SPLEED).

Recently SPSEE has been applied to study the temperature dependence of the magnetization in thin films in the spin-wave regime[150] and for thin Fe layers in the critical regime.[145] Also, SPSEE gives information on the reduction of the average magnetization upon adsorption.[149]

5. Spin-polarized electron energy-loss spectroscopy (SPEELS)

It has recently become possible to measure spin-dependent electronic excitations in ferromagnets by spin-polarized electron energy loss spectroscopy (SPEELS). SPEELS has been applied to Ni, Fe, and Co surfaces.[151,152] When in addition to using a primary polarized beam the polarization of the scattered electrons is also measured, an unambiguous deconvolution of the scattering processes into "flip" and "non-flip" channels is achieved. It was shown that for Fe and Ni exchange scattering constitutes a significant part of the total energy-loss processes. These data also show that the energy-loss rate in ferromagnets can be very spin dependent. In particular, the energy-loss probability for spin-down electrons in Ni can be four times higher than for spin-up electrons. These findings have certain bearing on the spin dependence of the electron mean-free path in ferromagnets and provide a possible explanation for the polarization enhancement and surface sensitivity in spin-polarized secondary electron spectroscopy. The probing depth (5 Å in $3d$ metals) might be determined by the mean-free path for inelastic exchange scattering. In other systems (rare-earth metals) the situation is very unclear. Measurements on rare-earth overlayers seem to indicate very strong exchange scattering leading to a probing depth of only ~1 monolayer. It is obviously very important to understand the spin-dependent scattering mechanisms in these systems in order to interpret secondary-electron polarization and polarization in other types of spin-polarized spectroscopies (e.g., photoemission).

The SPEELS experiment with polarization analysis does not require a ferromagnetically aligned sample, since spin-flip transitions can still be detected by a change in the polarization (depolarization). This is equivalent to polarized neutron scattering where, for example, magnons above T_C can be detected. Recently SPEELS on Ni above T_C revealed inelastic spin-flip scattering, as shown by a strong depolarization. This is evidence for the existence of local moments in the paramagnetic state and the persistence of a spin-split electronic structure[151] above T_C.

Since no long-range ferromagnetic order is required, SPEELS can also be applied to other systems, like antiferromagnets. It may be possible to measure the exchange splitting on a Cr(100) surface, which is predicted by theory to be on the order of 2 eV because of the large enhancement of the magnetic moment (2–2.5 μ_B) at the surface. The energy resolution currently achieved in SPEELS can be improved to less than 10 meV. This would open up the field to study collective spin excitations (magnons) at surfaces by SPEELS.

6. Polarized particle probes

In addition to the above spin-polarized adaptations of conventional electron spectroscopies, there are additional polarized-particle probes of surface magnetism. These utilize spin-polarized atom,[153] ion,[154] or positron[155] beams. The atom and ion beams are sensitive to the outermost layer of the surface region. They consist of spin-polarized metastable helium He(2^3S) and grazing incidence (~150 keV) polarized deuterons, respectively, which impinge on a magnetized surface. The de-excitation of the atom beam involves interatomic Auger processes. The emitted electrons have an asymmetry which depends on the spin orientation of the probe atom with respect to the magnetization of the target sample. In Electron Capture Spectroscopy (ECS)[154] the deuteron neutralization is detected by a nuclear reaction that yields ^{4}He particles whose angular-distribution asymmetry, caused by hyperfine interactions, provides a measure of the spin polarization of the captured electrons. In both spectroscopies the detected asymmetries can be studied as a function of temperature, crystal face, chemisorption, etc. to obtain surface-magnetism information. ECS has been used quite extensively to address many of the major issues in the field; on the other hand, spin-polarized metastable-atom de-excitation spectroscopy, like polarized positron scattering, has been demonstrated only in feasibility studies.

C. Electron microscopy

Electron microscopy offers the opportunity to characterize both the magnetic structure and the atomic structure of materials in the same optical column. Its spatial resolution is its most significant advantage, and the magnetic behavior of a material can be directly related to heterogeneities of both structure and composition on a near-atomic scale. Direct observation of domain wall pinning is possible, for example, and the atomic structure of the pinning site can be completely determined. There are no new requirements imposed on samples for imaging of their magnetic structure (standard microscope specimens can be used), although a change in operational mode of the microscope is essential, as described below.

Characterization of the localized magnetic structure of a sample includes the direct determination of magnetic domain size and morphology, the structure of

domain walls, the location and strength of stray fields, and the magnetization direction of all magnetic features. It might also include the dynamic observation of how the magnetic sample responds to the application of externally applied fields, increasing temperature, mechanical stress, and the like.

Characterization of the localized atomic structure of a sample includes its local crystal structure and orientation, grain size and morphology, defect structures (including dislocation, stacking faults, twins, grain boundaries, voids, and inclusions), second-phase particles (including their structure, composition, and internal defect structure), compositional variations (e.g., segregation at internal interfaces), and the atomic structure of surfaces and interfaces.

To conduct these studies in the electron microscope involves operating the imaging systems in a way that is sensitive to the localized magnetic fields within the sample, and comparing the resulting images to the more traditional images formed by scattered electrons. More recently, the use of a detector that is sensitive to the spin polarization of the electrons has been utilized for imaging domains.[156]

1. Scanning electron microscopy with polarization analysis (SEMPA)

For many purposes, it is desirable to have a high-resolution domain imaging technique in which the contrast is proportional to the magnetization, as in imaging by the magneto-optic Kerr effect, but not be constrained by the resolution limitation imposed by the wavelength of light. Further, one wants an imaging technique that can be applied to thick specimens in order to image magnetic structure on a nonmagnetic substrate, such as a bit written on a magnetic disk or a permalloy memory element on a silicon chip. This would avoid the need for thinning the specimen, as required for Lorentz microscopy on the transmission electron microscope (TEM), which is not only tedious but can change the magnetic properties to be studied. Such a high resolution imaging technique[157,158] is realized in SEMPA. By measuring the spin polarization of the same secondary electrons which form the Scanning Electron Microscope (SEM) topographic image, one simultaneously obtains an image of the magnetization with the high resolution of the SEM. All three components of the magnetization can be measured. Because of the inherent inefficiency of currently available spin analyzers,[156,159] the polarization measurement takes approximately 10^4 times as long as an intensity measurement of comparable precision. The resolution at present is 40 nm. In the near future sub 10 nm SEMPA resolution is expected for an SEM with a field-emission cathode. The secondary electrons sample at most the outer few nanometers of the specimen so that SEMPA is sensitive to the magnetic microstructure at or near the surface. This is an extra advantage for studying surface and thin-film magnetism.

An example of a SEMPA image[160] of a test pattern written on a thin film hard disk is shown in Fig. 9. The magnetic material is a 70 nm thick film of approximately 80% Co–10% Ni. A low magnification image is shown in Fig. 9(a). The light and dark stripes oriented approximately horizontally comprise the test pattern of written domains or bits. The bits were written successively in tracks, seven of which are seen running vertically. Domains of antiparallel magnetization appear as alternating black and white areas. The magnetization lies in the plane of the film, as indicated by the arrows in the higher magnification image shown in Fig. 9(b). The three nearly complete horizontal bands in Fig. 9(b), two dark and one light, are domains which at this magnification are seen to have irregular boundaries. Information is associated with the transition from one domain to another. A sharp, well-defined boundary is desirable for the minimum noise signal (see Sect. V, Applications). The jaggedness of the domain boundary clearly puts a limit on the maximum recording density. At the same time that one measures the components of the magnetization, one also obtains the conventional topographic image from the secondary-electron intensity, as shown in Fig. 9(c). This intensity image is for the same area as the magnetization image of Fig. 9(b) and shows the grooves commonly found on a hard disk. The SEMPA magnetization image is independent of the topography, which is an advantage relative to Kerr or Lorentz microscopy magnetic imaging where topographic and magnetic contrast can be difficult to separate.

2. Lorentz electron microscopy

Lorentz microscopy exploits the Lorentz force exerted on the imaging electrons by the internal and stray fields associated with a magnetic sample, and can be applied in scanning or transmission modes. Recalling that in the SEM the scattered electrons are collected by a detector that sits above the surface of the sample, the Lorentz force affects both the secondary-electron signal (particularly from the stray fields above the sample surface) and the backscattered electron signal (particularly from the influence of the internal induction of the backscattered yield). These two effects result in "Type 1" contrast and "Type 2" contrast, respectively, and have been used for some time.[161]

To obtain contrast from Lorentz-scattered electrons in the transmission electron microscope (TEM), they can either be blocked with an objective aperture (Foucault mode), displayed by phase interference through defocusing the objective lens (Fresnel mode), or exhibited in holographic fashion, also by phase interference.[162]

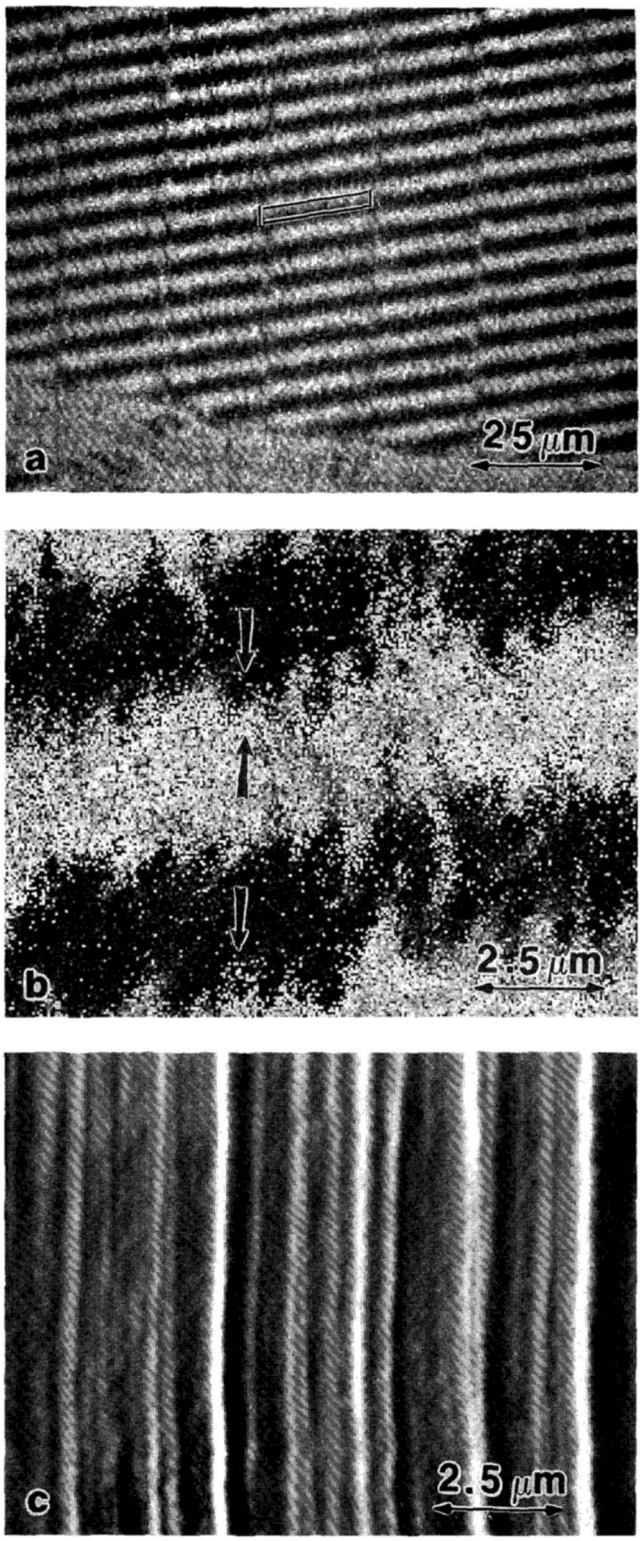

FIG. 9. The SEMPA image in (a) shows approximately horizontal light and dark bands corresponding to a written test pattern of magnetic bits, one of which is outlined near the center. In (b) a ten-times-higher magnification magnetization image than in (a) shows the irregularity of the domain boundaries which contribute to the read-back noise and ultimately limit the density of information that can be recorded. The intensity image in (c) shows the surface topography of the same region as in (b).[160]

The last technique shows lines of constant induction in the sample, but places severe restrictions on the thickness (and flatness) of the sample.

In the scanning transmission electron microscope (STEM), the Lorentz-deflected electrons are best detected in the far field, so that the signal at the detection plane is stationary, even though the incident beam is scanned. This can be done by methods similar to the TEM, or by the use of a split detector that forms an image on the basis of Differential Phase Contrast (DPC).[163,164] A display of the difference signal between segments of the split detector shows the regions of the sample that provide the Lorentz force on the imaging electrons. Because such a signal may have any direction within the plane of the specimen, it is important to be able to rotate either the sample or the detector. A more acceptable option is to divide the detector into many segments (quadrants at least) and scan through the detector signals until a difference image is detected. With electronic manipulation of the signals, a variety of micromagnetic information can be obtained, including the quantitative assessment of induction integrated over the electron path.

The most severe limitation on spatial resolution in Lorentz microscopy methods is the need for a magnetic field-free region around the specimen. Electron microscopes rely on electromagnetic lenses for resolution (probe-forming in SEM, aberration correcting in the TEM), with optimum resolution coming from strong fields in which the specimens are immersed to maintain short focal-length conditions. Obtaining a field-free condition requires that the microscope be run in long focal-length conditions, sometimes achieved by actually turning off the objective lens in the TEM or STEM. This unsatisfactory condition is being addressed with the design of new field-free lens configurations,[165] and by the use of field-emission guns for better probes during beam scanning. Nevertheless, the current resolution limit for Lorentz methods in electron microscopy is approximately 3 nm.

3. Conventional electron microscopy

The most attractive aspect of Lorentz microscopy is that it is readily complemented by conventional methods of microstructural evaluation, within the same instrument. By switching to normal imaging modes in the SEM, TEM, or STEM, microstructural information is rapidly obtained, at the superior spatial resolution of these methods. In TEM, atomic resolution is now achievable, and can be applied to image most magnetic materials, as long as the volume of magnetic material is not so large as to aberrate the imaging beam. Furthermore, complementary diffraction and spectroscopic methods popular in electron microscopy can also be applied. These include selected area diffraction, micro-

diffraction, convergent beam electron diffraction, energy-dispersive spectroscopy of x-rays, and electron energy loss spectroscopy. Spatial resolution in these methods is limited by spot size, which is currently in the 2 nm regime. In the SEM, crystallographic information can be obtained by electron channeling patterns, and morphological information from standard imaging procedures. Compositional maps that show local spectroscopic information at the 50 nm range are universally attainable.

It is still necessary in TEM and STEM methods to thin the sample to the condition of electron transparency, often with specific geometrical constraints so that the electron beam traverses an interfacial region in cross section, for example. Many methods are available for such sample preparation, and they can be applied to bulk, thin film, or multilayer magnetic materials with no modification. Even if exact geometrical orientation is missed during sample preparation, the microscope goniometer can usually be used to adjust the orientation during imaging. Finally, the use of computer modeling of the image formation process lends credibility to the interpretation of even the most complex images, and the current trend is to have such simulations available on-line.

4. Scanning tunneling microscopy (STM) and magnetic force microscopy (MFM)

In the last few years the STM has emerged as a powerful means to study surface structure at the atomic level. Rather spectacular results have been obtained for semiconductor- and metal-surface reconstructions and for adsorbates on such surfaces. So far the STM has not been used significantly to characterize growth, although it has the potential to answer such an important question as whether a uniform, continuous monolayer of material, a prototype two-dimensional metal film, has been achieved. There is a great opportunity to correlate STM results with those of other techniques in efforts to characterize more fully growth of magnetic thin films.

A natural question is whether it is possible to extend the STM to include spin sensitivity so that spin configurations can be imaged with atomic resolution. The possibility of using a magnetic tunneling tip, such that the tunneling electrons are polarized and must find empty states of the same spin to tunnel into, has been considered.[166] Several hurdles to achieving this spin sensitivity have also been considered. The magnetic electrons, for example *d* electrons, are more localized than *s-p* electrons and thought to tunnel about a hundred times less efficiently. Furthermore, there are expected to be strong interactions between magnetic tips and samples, such that the spin configuration to be measured could be significantly distorted. Ideally one wants to control the polarization of the tip electron and reverse it at will. An optically pumped GaAs tip may overcome some of these problems, but formidable effort will be required to implement the technique.

The MFM uses a fine magnetic tip on a cantilever of small spring constant to detect variations of the magnetic field or field gradients just above the surface.[167] The MFM suffers from the same problem as a magnetic tip STM in that there is a perturbing interaction between tip and sample.[168] It does not have the high resolution gained by tunneling; it is limited by tip size, and lateral resolutions of 1000 Å may be expected. The MFM is in many ways similar to the Bitter technique. It has the advantage that it can operate in air, and it senses the stray magnetic field which is the information wanted for some magnetic applications. It does not appear to be well suited for studies of domain wall structures or to obtain information on the sample magnetization.

D. Diffraction

Conventional diffraction techniques have been used for many years to determine the structure of bulk materials. As such, these techniques are well established and therefore can be used reliably. The application of diffraction techniques to thin films, surfaces, and interfaces is limited by the small amount of material available in the sample and the complications caused by the presence of substrates. These difficulties, however, can be overcome by the use of more intense radiation sources, and by a more detailed understanding of the structure of the substrate.

1. X-ray diffraction

It is quite clear that the magnetism of surfaces, interfaces, and films is intimately connected with their physical structure. Therefore, magnetic studies are of doubtful validity in the absence of complementary structural information. While neutrons, x-rays, and electron diffraction can play useful roles, x-ray analysis is the best established and probably the most powerful probe of overall structural characterization. Well established techniques exist to deal with structures of great complexity, with defects, and with structural rearrangements. Furthermore, new synchrotron x-ray sources provide enormous intensity, thereby opening new avenues to the x-ray study of chemical, and even magnetic, structure.

Determination of the structure of interfaces in thin film systems is a difficult problem requiring the development of new methodologies and the refinement of older ones. Defects such as roughness, interdiffusion, and dislocations become major determinants of the diffracted intensity. It is precisely the same structures that

dominate, in many cases, the magnetic properties. Although the problem has received considerable attention,[42,169,170] most studies begin with a model of the disorder, include it in a structure-factor calculation, and compare the results with experiment. For example, the Fresnel formalism has been used extensively to interpret small-angle diffraction data from multilayered films,[171,172] with roughness parameters introduced phenomenologically through a pseudo-Debye-Waller factor. For true superlattices (crystalline films), on the other hand, standard kinematical theories have been applied to treat the effect of disorder on large-angle diffraction peaks.[173] While these theories give qualitative predictions, a quantitative understanding is lacking. In systems where large- and small-angle data are both available, for example, it has not been possible to produce a model that brings the two sets of data into quantitative agreement.[174]

Recently a nonlinear optimization method, similar in spirit to the extensively used Rietveld refinement method, has been applied to x-ray diffraction data from films and multilayers.[172] In this approach, continuous and discrete roughness, both perpendicular to, and in the film plane, interdiffusion, and polycrystallinity are included as adjustable parameters for refinement. Very high quality fits can be obtained in this fashion for diffraction data from superlattices. An alternative approach uses a diffraction model for multilayers, decreasing higher Fourier components of the composition and lattice parameter variation by adjustable damping factors.

Before these techniques become standard, well controlled experiments should be performed on samples with induced, controlled disorder. The results should be compared with neutron-diffraction and electron-diffraction results. The latter are particularly important in helping to distinguish local random roughness, such as caused by fluctuations in growth conditions, from correlated roughness caused by systematic drifts in preparation conditions.

2. Neutron scattering

Although neutron scattering can give, in principle, the same structural information as x-rays, it suffers from lower intensity and resolution and is, of course, tied to major facilities. At the present time, however, neutron scattering is the method of choice for determining magnetic structure and, especially, detecting magnetic excitations. The magnetic cross section for neutron scattering, while small, still permits determinations of thin-film and multilayer magnetic structures. Conventional triple-axis methods, with moderate neutron fluxes, have been used successfully to determine the detailed magnetic structures of Dy/Y and Er/Y multilayers,[175] and of Er films[61,110] as thin as 30 nm ($\sim 10^{-6}$ cm^3 of Er). By increasing the area of the film from 1 cm^2 to 10 cm^2 and working at the highest flux currently available, it should be possible to extend such measurements to the 1–3 nm regime. Substrate background becomes a major factor, requiring energy analysis and a reduction of substrate volume to enhance this. Polarization analysis increases sensitivity further by separating magnetic and nuclear scattering and is particularly important for ferromagnetic films. This has been especially useful in studies of Gd/Y and other multilayers.[62,107]

Analysis of magnetic neutron data requires simultaneous knowledge of the chemical structure. In the Dy/Y and Er/Y work, the structure was modeled by a Fourier series for a square wave with damping of successive terms.[39] Simultaneous treatment of nuclear and magnetic peaks with different damping factors for composition, lattice spacing, and magnetic modulation permitted a layer-by-layer determination of magnetic moment and orientation in these modulated magnetic structures. Similar procedures can be used to model the strain distribution in thin films if data are taken at numerous reciprocal lattice points.

Techniques are becoming available to probe the depth dependence of the magnetization in thin magnetic films or at the surface of bulk magnetic systems by neutron scattering. This technique, Polarized Neutron Reflectometry (PNR), was developed by G. P. Felcher at the Argonne National Laboratory and involves reflecting spin polarized neutrons at grazing incidence from the surface of the specimen.[176] The reflection of the neutron beam can be described by a spin dependent, depth (z) dependent refractive index of the specimen,

$$n^{\pm}(z) = 1 - c\{b \pm B(z)\},$$

which includes contributions from the nuclear and magnetic neutron scattering. For typical materials n differs from unity by 1 part in 10^5, which gives critical angles for total reflection of less than one degree. The experiment consists of measuring the reflectivity of spin up and spin down neutrons at a fixed incident angle as a function of neutron wavelength for wavelengths up to those for which the neutrons are totally reflected. The reflectivity, calculated from models of $n^{\pm}(z)$, is compared to the experimental reflectivity curves.

The first use of PNR in magnetic materials[177] was to determine the magnetization profile of a sputter-deposited film of Fe_3O_4 approximately 2500 Å thick and to compare it to that of the same film after further oxidation to γ–Fe_2O_3. Surprisingly, the reflectivity data on the annealed film showed that there was a nonmagnetic layer at the surface about 150 Å thick. Later the magnetic inhomogeneity of the film was shown to arise from the formation of a nonmagnetic α–Fe_2O_3 phase at the surface of the film. The distribution of the

nonmagnetic phase through the film, as determined from grazing incidence x-ray diffraction, was shown to be in very good agreement with the detailed variation of magnetization deduced from the PNR data, providing confirmation of the power of the PNR method.[178]

The primary merits of the PNR technique are that it is nondestructive and gives the absolute magnitude of the magnetization for depths up to about 5000 Å from the surface. There is no lateral resolution in PNR, and it requires very flat samples about 1 cm^2 in area. However, PNR can probe magnetic layers buried beneath several hundred angstroms of nonmagnetic or antiferromagnetic layers. For example, recently the magnetic structure of permalloy layers comprising part of an exchange coupled structure of the form Si(111)/NiFe (400 Å)/FeMn (400 Å)/ Ta(200 Å) has been successfully probed[179] using PNR. It is thus established that this technique is able to probe buried magnetic interfaces. At present the depth resolution of PNR is limited to about 20 Å. However, in recent experiments it has been shown that it is possible to determine the magnetization of ultra-thin layers of Fe and Co, enhancing the spin asymmetry of the neutron reflectivity by covering the magnetic layer with a thin layer of Cu, which acts as an antireflection coating.[180] The enhancement of the spin asymmetry of $R\pm$ depends sensitively on the thickness of the overlayer. With this method the magnitude of the magnetic moment of Fe and Co layers only one monolayer thick was determined.

Since PNR is sensitive only to the component of magnetization normal to the scattering plane, by orienting the magnetization of the sample perpendicular to the sample plane the neutron reflectivity becomes spin-independent. It is then possible for the composition of the sample to be determined as a function of depth from the surface in the same experiment. Such detailed information is required in modeling the spin-dependent reflectivity curves to obtain the magnetic structure of the film. Finally, the depolarization of the reflected neutron beam gives information on any lateral inhomogeneities in the sample, such as those resulting from formation of magnetic domains of a certain size.

A promising extension of PNR now under development[181] locates a second detector at a fixed angle above the reflection plane. By varying the incident neutron wavelength and sample orientation it should be possible to perform surface neutron diffraction from surface magnetic structures. The scattering wave vector in this case lies in the plane of the sample, making this probe sensitive to moments oriented normal to the sample surface. Surface antiferromagnetism and surface magnetic reconstruction may be detectable by such techniques.

Because of the very high energy resolution ($\leqq 1$ μeV) possible, studies of dynamical processes in solids have long been dominated by neutron scattering. So far, studies of quasielastic scattering (critical phenomena), spin waves, and magnetostatic modes in thin films have not been reported, but should be possible with the combination of larger sample areas, the use of multiple, identical samples, and increased neutron flux. A serious problem here (as in elastic scattering) is the strong scattering from the substrate on which the film or multilayer is grown. Triple axis methods can eliminate inelastic scattering from the substrate for structural determination, but only elimination or reduction of the substrate scattering will suffice in inelastic studies. Such studies will complement optical techniques (which are restricted to small momentum transfer) and electron scattering (which may suffer from multiple-scattering effects).

E. Photon sources

The major new developments in synchrotron-radiation sources open new research horizons in novel magnetic-film studies. At present the first spin-polarized photoemission initiative in the United States has been established at an undulator beamline on the VUV ring at the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory.[136] The superior flux and brilliance of the undulator source helps to compensate for the inefficiency of the spin detector. These characteristics permit magnetic materials studies to proceed systematically, whereas the earlier European efforts on bending-magnet beamlines were primarily valuable to demonstrate the feasibility of the approach. It is expected that spin-polarized band mappings will result from the synchrotron efforts; the results will test local-density functional calculations of the electronic structure of surfaces and metastable epitaxial phases.

Future developments involve the forthcoming availability of the Advanced Light Source (ALS) at the Lawrence Berkeley Laboratory,[182] which will provide additional undulator-beamline capabilities to satisfy the expanding needs of the growing community of novel magnetic-material researchers. A somewhat more speculative advance would involve the availability of free-electron laser (FEL) sources in the VUV/soft-x-ray range.[183] Such sources are being conceptually designed at present.[184] The photon-energy tunability is comparable to that of synchrotron-radiation (SR) sources, but the intensity, brilliance, coherence, and monochromaticity are all projected to surpass substantially the performance of SR sources. The several orders-of-magnitude increase in intensity would permit magnetism researchers to perform analogous experiments to those envisioned by other materials researchers with undulator sources.[185] These include pump-and-probe experiments to study magnetic excited states by synchronizing conventional laser sources with the FEL pulse train. Also, spin-polarized photoelectron micros-

copy can be envisioned, which would benefit from the superior brilliance of the FEL. (The high intensity of the FEL source, in addition to providing new opportunities, also raises the problematic issue of space-charge effects.) The monochromaticity would permit the spin asymmetry at the Fermi energy E_F to be obtained for comparison with transport and susceptibility studies. Only at E_F can the full potential of the FEL-source resolution be realized, because at E_F there are no Auger processes to introduce lifetime-broadening effects into the spectroscopic results. Still, it is interesting to consider whether enhanced resolution would enable effects such as magnon sidebands to be observed on core-level spectra.

Core-level spectroscopy will benefit also from the Advanced Photon Source (APS) at Argonne National Laboratory,[186] which is projected to be operational in the late 1990s. Spin-polarized core-level analysis has recently been proposed as a means to monitor short-ranged magnetic order, and is applicable to antiferromagnets as well as ferromagnets.[187] Conventional photoelectron diffraction from core-level emission also provides an advanced structural-characterization tool for epitaxial monolayer-type structures.[44] The APS will provide much needed structural characterization capabilities as well, through the use of grazing incidence surface-structure analysis. This is the technique in which x-ray crystallography is performed in the total-external-reflection geometry to enhance surface sensitivity.[188] The value of magnetic x-ray scattering to understand the bulk magnetic structure of the heavy rare-earth spiral spin arrangements has also been demonstrated.[189] The technique has also been proved to be effective in characterizing magnetic superlattices.[190] It would be fascinating to combine magnetic x-ray scattering with the grazing-incidence geometry to obtain surface magnetic structures. These studies require the anticipated brilliance of the APS undulator beamlines, and cannot be performed at existing hard x-ray sources. Another challenging possibility involves the ability to separate the spin and orbital contributions to the magnetic form factor by means of x-ray scattering.[191] It has been demonstrated, in principle, that such a decomposition is possible, although experimental confirmation is yet to be achieved. Since the role of the spin-orbit interaction is so seminal to understanding the surface anisotropy and the magneto-optic response, any additional information on spin-orbit effects is very welcome. Such studies would benefit as well from the availability of circularly polarized x-rays, because of their enhanced magnetic-scattering cross sections relative to linearly polarized x-rays that are more commonly produced at synchrotron sources. In summary, the combined approach of using advanced synchrotron sources to obtain structural, magnetic, and electronic-properties information provides extraordinary research opportunities for future research in the field of novel magnetic materials.

V. APPLICATIONS

Many of the applications of magnetic-material systems require control of the extrinsic properties such as coercivity, orientation, permeability, and micromagnetic features. Intrinsic properties such as moment, anisotropy, or magnetostriction are normally accepted as given for a particular material composition. In the last few years it has become clear that a better understanding of the interactions at the interfaces of materials can be used not only to control the extrinsic properties but to manipulate the intrinsic properties as well.

While not all of the preparation and characterization procedures described earlier are the methods of choice in a manufacturing environment, the fundamental understanding of the role of the interface developed by the use of these techniques should provide a guide in selecting materials and process conditions for commercial applications.

The following are only a few examples of applications of magnetic materials and systems. In each of these the ability to improve performance significantly requires an understanding of the interface interactions so that new material systems and practical processes can be developed. It is worth noting that many of the fundamental concepts are common to many of the applications. For instance, the magnetic anisotropy and the exchange interaction across the interface directly influence the coercivity of both hard and soft magnetic materials.

A. Magnetic recording

Currently there are many different recording media for recording systems. While videotape systems operate at similar track widths to high-performance hard-disk systems—typically 1000–1500 tracks per inch (tpi)—the linear densities are greater by factors of two to four—80 kiloflux changes per inch (kfci) versus 30 kfci. This difference can be justified by the error rates required and data encoding schemes used. If the performance-improvement-versus-time curves that have characterized the last thirty years are to continue, the area densities will increase by a factor of twenty by the turn of the century. This will require not only that the mechanical interface between the magnetic head and the media be improved but also that magnetic recording properties of the media be much improved. For example, a longitudinal recording system with trackwidths of less than 2 μm (10 000 tpi) and linear bit densities of 100 kfci will require high coercivities ($\geqq$2000 Oe) and very smooth media so that extremely close head-medium spacings ($\approx$500 Å) may be ob-

tained. At these small sampling volumes of a bit, medium noise, head noise, and head output become major magnetic concerns.

1. Hard disk media

Current laboratory media for hard disk thin films are composed of a substrate upon which a series of layers of materials are deposited. Typically a nonmagnetic underlayer, such as Cr, is sputtered by means of a dc magnetron over an amorphous Ni–P layer which has been electroless-plated on an aluminum disk. A number of Co-based alloys, such as CoNiCr, CoP, CoCrTa, or CoCrPt have been used as the sputtered magnetic layer over the underlayer. Over this layer a nonmagnetic, thin overcoat is deposited to protect the media during the time the head comes in contact with the surface. The dominant philosophy is to achieve in-plane anisotropy, high coercivity, and low noise. The efforts are thus centered around control of interfaces. The nonmagnetic underlayer, Cr, is deposited with selected thicknesses; film growth conditions are chosen so as to provide grain size control and particular crystal planes are exposed for the magnetic layer to grow on epitaxially during sputtering. This underlayer provides an interface by means of which the hexagonal-close-packed cobalt alloy can be oriented in the plane. The anisotropy of the alloy provides a coercivity mechanism if the grain size is appropriate for a single domain per grain. The underlayer grain size can be replicated by the magnetic layer. The magnetic alloy and deposition conditions are such that during deposition various phase segregations[192,193] at the individual grain-grain interfaces can occur. If these interfaces sufficiently decouple the magnetostatic and quantum-mechanical exchange interactions between grains, then the roughness of the recorded bit edge will essentially be defined by the geometry of the grains. The medium noise will then be determined by grain-counting statistics, just as in good particulate media, and not by arbitrary zigzag-shaped domain wall boundaries.[194,195]

The very small future bit volumes will require much smaller grain sizes (currently about 1000 Å) so as to minimize medium noise. In order to maintain signal output, the volume of the nonmagnetic decoupling intergranular boundaries should be reduced so that the total magnetic moment per bit is as high as possible. Fundamental understanding of interface decoupling layers would be helpful, in this context, to guide material selection. To achieve high coercivity within these very small particles, a better understanding of the surface anisotropies that can be induced by the surface of the nonmagnetic underlayer may be essential. In even more futuristic media, as the grain size approaches the superparamagnetic limit, surface anisotropy may be needed to provide the required domain stability.

2. Magneto-optic media

After many years of development, magneto-optic drives have recently reached the market place. These drives use a medium composed of an amorphous rare-earth transition-metal alloy (e.g., Tb–Fe–Co). The information is recorded by localized heating with a laser and switching the magnetic state. Readout is accomplished by a combination of the polar-Kerr and the Faraday effects. Recording density performance of the system is largely determined by the spot size of the laser ($\lambda \sim 800$ nm). Bit cell sizes are 1.5 μm to 2 μm on a side. The signal-to-noise ratio is largely determined by the size of the magneto-optic effect and the optical depolarization caused by medium imperfections. The carrier-to-noise ratio (30 kHz bandwidth) is typically better than 50 dB.

Since the recording densities of current systems are restricted by the diffraction limit of the light, improvement in future systems will require materials which have large magneto-optical effects at shorter wavelengths ($\lambda < 500$ nm). The key issues are to maintain the perpendicular anisotropy, control the Curie temperature, have a reasonably high magneto-optic effect, and obtain a noiseless, nondepolarizing grain structure. Over the years a considerable number of materials with larger magneto-optic effects than the amorphous rare-earth-transition-metal alloys have been investigated. Almost all, however, suffer from depolarization noise.

Recently, effort to make new magneto-optic systems with compositionally modulated films has shown some promise.[196] These films should have very little grain noise. A few angstroms of cobalt layered with several angstroms of palladium or platinum have been shown to possess perpendicular anisotropy; thicker films do not. The optical effects at short wavelengths show promise, but need improvement. Increasing the ratio of Co to Pd or Pt would help the optical properties but degrade the anisotropy. In order to make films of this type useful for future magneto-optic recording systems, an improvement of the interface-induced anisotropy and a better understanding of the attendant mechanisms are needed.

3. Exchange biasing of magnetoresistive heads

Recent years have seen increasing use of the magnetoresistive (MR) read head as a replacement for the traditional inductive head in the reproduction of magnetic recordings. Advantages include a velocity independent signal and lower head noise in some applications. The MR head uses the magnetoresistive effect to change the voltage across a thin-film element placed near the recording medium and its accompanying fringe fields. This element, typically FeNi, is usually biased by an external field so that it operates in a linear regime and consists of a single magnetic domain. The external field

may be provided by a neighboring permanent magnet; however, exchange biasing through an antiferromagnet (typically FeMn or TbCo) is receiving increasing attention.[92,197] The major discrepancy between experimental exchange biasing and simple theory limits the adjustment of the biasing for engineering purposes. Thus, a quantitative theory of exchange biasing would be highly beneficial for this application.

4. High magnetization materials for recording heads

As the recording medium coercivity is raised to increase recording densities, it becomes necessary to produce larger fields with the recording head. Saturation effects, unfortunately, limit the field which may be obtained from a head. Hence, new magnetic thin film materials with higher saturation flux densities than Permalloy (B_s = 10 000 G) are needed. In addition to offering high saturation flux density, these materials must also have zero magnetostriction and be resistant to annealing during processing. There has been considerable work on amorphous metal-metal systems (e.g., Co–Zr) and amorphous metal-metalloid systems (e.g., Fe–Co–Si and Fe–Co–B). These materials offer saturation flux densities of up to 14 000 G in nonmagnetostrictive compositions. There has also been work on iron nitride materials, with theoretical saturation flux densities as high as 25 000 G, although nonzero magnetostriction and stability of the materials during annealing are serious limitations. There have been attempts to develop multilayers with high saturation flux density. Multilayers of Co–Zr/Fe, in which the Fe is kept amorphous by depositing in extremely thin layers, have been produced with saturation flux densities of 18 000 G. Again, nonzero magnetostriction, which changes with annealing, is a serious problem. Another future possibility is that a suitable growth technique could be found either to fabricate superlattices which take advantage of the enhanced magnetism at interfaces or to produce controlled strain effects which increase the magnetization. An additional possibility could be to employ single-crystal materials, grown by MBE or simpler techniques, which could have a reduced coercivity caused by the absence of pinning states for the domain walls.

B. Magnetoelastic devices

Applications of magnetoelastic phenomena in thin-film devices may exploit two classes of magnetic materials. The first are amorphous transition metals, with nearly zero anisotropy, which exhibit the highest magnetomechanical coupling factors ever observed; thus they can be used as ultra-sensitive magnetic-field detectors with a displacement readout, or strain detectors (accelerometers, etc.) with an inductive readout. These materials can be sputtered in thin-film form. A hybrid piezoelectric/magnetoelastic structure can be envisioned which would convert voltage to magnetic field and vice versa.

The second class of materials are rare-earth transition-metal alloys, which have more moderate magnetomechanical couplings but produce large strains/high power at reasonable magnetic fields. Such materials might be used in thin-film form or in a superlattice composite to control the state of strain in a nonmagnetic material (e.g., semiconductor).

C. Integrated optical and electronic devices

Although there has been considerable activity in the growth of thin magnetic films, there has been little effort to introduce magnetic elements into either integrated optical or integrated electronic circuitry. There are, nevertheless, many opportunities for such elements to provide nonreciprocal devices to act as isolators, phase shifters, delay lines, circulators, or filters. All of these devices are used in current high-frequency technology (microwave signal transmission, radar, etc.). As the need develops for ever higher frequency operation, the shorter wavelengths require that the dimensions of the devices shrink. These devices become so small that they must be monolithically incorporated into the integrated circuits on a microchip. Thin magnetic films are, therefore, the appropriate morphology for this new technology. One can either use magnetic insulators to act as a dielectric medium within strip-line devices, or magnetic metals to act as either guidelines or ground planes for the devices. In either case the challenge is to provide magnetic films on appropriate substrates (Si, GaAs, etc.) having the appropriate magnetization and anisotropies.[198]

A second use for magnetic materials is to provide a magnetic field on an integrated circuit substrate. There are many devices which require an applied field, but it is impractical to attempt to house a microchip actually within the field of a coil, electromagnet, or permanent magnet. What is required is the means of providing, on a very small scale (≈ 1 μm), a highly localized magnetic field which affects only a single circuit element. Furthermore, this magnetic field must be provided by a magnetic material, which can be laid down in film form and readily patterned by techniques which are compatible with other fabrication techniques common to the semiconductor microchip industry.

Surfaces and interfaces play an important role in these applications in determining both magnetization and anisotropy of the films. For example, as mentioned above, chemical reactions can diminish the magnetization at the interface. Appropriate buffer layers at the interface can prevent these reactions. The nature of the

growth at the interface can often introduce oriented strains or dislocations which lead to anisotropies. These anisotropies may be useful in providing easy axes for the magnetization or could prove troublesome if one requires isotropic behavior in the film. Examples such as these illustrate the importance that details of the film growth can have on the ultimate application.

D. Permanent magnets

It has been estimated that by the mid-1990s the commercial market for the rare-earth-transition-metal alloys as materials for permanent magnets will be several billion dollars annually.[199] The discovery and rapid development of the $Nd_2Fe_{14}B$ class of materials is making this possible. The emphasis on applications of the relatively inexpensive $Nd_2Fe_{14}B$ has provided increased awareness of $SmCo_5$ and Sm_2Co_{17} materials and is increasing the demand for them. Special batches of $Nd_2Fe_{14}B$ with energy products approaching 50 MGOe have been made in the laboratory, while materials with values approaching 30 MGOe are becoming commercially common. Traditional ceramic ferrite magnets have energy products of approximately 4 MGOe. The extremely high energy product of the $Nd_2Fe_{14}B$ materials has allowed the size and weight of devices to be significantly reduced.[199] For instance, a 100 hp motor which normally weighs 1000 lbs can now be reduced in size to weigh about 35 lbs by using these magnets. Similarly, the electrical efficiency of a small fractional-horsepower motor can be doubled from 35% to 70%. The high coercivities are allowing significant improvements in the package design of planar or pancake-shaped motors and actuators.

These materials are manufactured either by a sintered, powder-metallurgy process or by a new rapid-quenching, hot-pressing, and die-upsetting process. Both processes result in very good products. A comprehensive review article on the rare-earth-transition-metal magnets is available.[199] In it, much of the current understanding of the materials and some of the uses for these magnets are described.

The principal issues for obtaining high-energy products are very similar to those described for the hard-disk thin-film media. The magnets need to be oriented in order to utilize their high magnetization. They need to have isolated single-domain grains along with a very high anisotropy in order to have coercivities higher than their $4\pi M_s$ values. The interfacial boundaries and the coupling between grains determine whether or not domain walls can be nucleated at, or propagated across, the interface. It is believed that nonmagnetic phases must be formed at these interfaces. However, the exact composition and role of these interfaces and phases are not clear. The temperature dependence of the coercivity is very important for many of the applications, and the role of temperature on the interface is an open issue.

VI. ISSUES AND PROSPECTS

This section is an attempt to highlight what the authors consider are important issues, and what they believe are the prospects for future research. Open and unsolved problems, current investigations and future prospects are mentioned throughout this report; some of them are reiterated here briefly. The goal is to bring together, in one section, a brief summary of research opportunities in surface, interface, and thin-film magnetism.

A. Theory

Ab initio techniques based on the local spin-density approximation have been very successful in predicting trends in magnetic properties; they need, however, to be applied—as resources and computer capabilities permit — to more magnetic-surface and thin-film systems. Details of surface magnetic properties which depend on the lowered symmetry require techniques which explicitly represent the solid-vacuum interface by means of boundary conditions on the electron wave functions or density—the so-called film codes. These calculations are extremely time-consuming and expensive, and the codes are in use at only very few institutions.

Magnetic properties of interfaces between two different materials, which also involve lowered symmetry, can be simulated either by film codes or by layered supercell calculations which use bulk codes. In either type of calculation the spatial scale of properties which can be investigated is limited by the number of atomic layers which can be included in a unit cell, currently of the order of 10. For example, investigation of the very interesting coupling observed between Fe layers in Fe/Cr/Fe sandwiches and Fe/Cr superlattices, which occurs for Cr thicknesses of 10–20 Å, will require a very large expenditure of state-of-the-art supercomputer time.

Structural relaxation at surfaces and interfaces makes the limitations of supercomputer resources even more acute, as the existence of such relaxations requires significantly larger unit cells. In addition, if structural relaxations are not known, calculations of many different structural configurations may be required in order to determine the minimum-energy relaxation. The greatest opportunities in this area of *ab initio* theory lie in the development of more efficient codes for film and layer calculations, in the development of algorithms for efficient searching of the phase space of structural relaxations, and in the careful choice of prototype magnetic systems for study.

The total-energy capabilities of *ab initio* calculations can be used in a different way to predict magnetic properties of thin films which are caused not by the lowered symmetry but rather by strain or lattice distortions due to interface bonding. Bulk codes have proved to be very successful in predicting the systematics of magnetic structure—nonmagnetic versus ferromagnetic versus antiferromagnetic arrangements—as a function of lattice parameters and lattice symmetry. Calculations have been performed for most of the 3*d* magnetic transition metals in the bcc and fcc structures. However, hexagonal and lower-symmetry structures—e.g., tetrahedrally, orthorhombically, or trigonally distorted bcc and fcc systems—are just beginning to be investigated. These types of calculation, which are relevant for epitaxially strained or "pseudomorphic" structures, are most useful for films sufficiently thin so that they remain pseudomorphic but thick enough so that the strain dominates the surface/interface effects. For these calculations to be extended to lower-symmetry systems, the approximations of spherical averaging of potentials or of electron densities used in the most efficient codes must be carefully evaluated, and perhaps eliminated in favor of full-potential codes that include nonspherically symmetric terms. Elimination of spherical approximations may also be crucial for achieving numerical accuracy in surface and interface calculations of the type discussed above.

Theoretical studies should also be pursued to determine the inherent limitations of local-density methods discussed in Sec. II (Theoretical Background). These limitations (the few percent errors in the lattice constants, for example) seem to be greater in spin-polarized magnetic systems. There is preliminary evidence that more complicated forms of the exchange-correlation potential, such as those that include terms in the gradient of the density,[200] may improve results in some cases.

In addition to the *ab initio* calculations, which are involved, expensive, and require state-of-the-art supercomputers, there is an obvious need to develop simpler correlations and empirical rules which could either provide qualitative explanations for existing experimental data, or point toward systems and configurations which might exhibit some required magnetic property. Attempts in this direction exist,[14,77] but they are still too crude to be of practical significance.

Beyond the calculations of equilibrium structures and primary magnetic properties (magnetic moment, hyperfine field, exchange splitting, etc.), there is a crucial need to determine secondary magnetic properties, such as anisotropy and magnetostriction, by means of electronic-structure techniques. These problems are discussed separately below.

Finally, the richness in structure and the complexity of the systems discussed here are, continuously, a source of surprises for new, unexpected, unexplained, or misunderstood effects which require both qualitative and quantitative explanation. Theory can develop only by the simultaneous paths of constant interaction between theory and experiment, and by the formulation of (by necessity) simple models able to extract, from the large number of secondary and irrelevant effects, the basic features of the phenomenon under consideration.

B. Magnetic moments at surfaces and interfaces

The values of the moments at the surface of magnetic metals remain a lively issue which needs more careful experimental data. All theoretical calculations[76–70] agree with the fact that at free surfaces the magnetic moments tend to be enhanced (in weakly magnetic metals) or created (in almost magnetic metals), even though the precise values of those moments tend to vary appreciably from calculation to calculation. The available experimental data, even though not 100% in agreement with each other, tend to confirm indirectly these theoretical predictions. Experimental confirmation, with direct experimental measurement of the specific surface and/or interface moments, is not yet available.

In particular, several issues require further clarification and reliable experimental data:

(1) The repeatedly calculated and indirectly observed large magnetic moment at the {001} surfaces of antiferromagnetic chromium—a ferromagnetic layer in an ideal, defect-free surface—remains yet to be observed *directly.*

(2) The optically observed (SMOKE) dead layers of iron [201] when deposited on ruthenium (0001) remains a puzzling effect which requires careful theoretical and experimental work. An accurate self-consistent calculation, including structural rearrangement effects, and the performance of the experiment at low temperatures is needed.

(3) The magnetic moments, if any, of the free surface of vanadium and of vanadium overlayers on a variety of substrates remain an open question.

These are only a few examples of systems which remain to be examined; the general area is still only sketchily explored and is a rich ground for basic research with possibly many practical applications.

C. Magnetic coupling at interfaces

One of the most exciting areas of both current and future research is that of coupled magnetic multilayered systems. In the simplest case, materials engineering of these magnetic systems allows for the optimization and control of such basic magnetic properties as saturation magnetization, anisotropy, coercivity, and mag-

netic domain structure. It seems clear that these types of structure will be of increasing importance in the magnetic-recording industry, as finer tuning of these magnetic parameters becomes necessary. At the opposite end of the spectrum, distinctly new properties of coupled magnetic systems have recently been discovered in a number of different magnetic systems. The most recent discovery is that of antiferromagnetic coupling of neighboring Fe layers in Fe/Cr/Fe sandwiches, together with an enhanced magnetoresistance in such systems.

Examples of areas of research likely to produce important new results include:

(1) A search for new ferromagnetic/metal/ferromagnetic layered structures displaying antiferromagnetic coupling of ferromagnetic layers. This search will likely lead to improved understanding of the phenomenon, and its related effects in magnetotransport properties.

(2) Study of the magnetic tunneling valve effect in ferromagnetic/insulating/ferromagnetic layered systems, and its dependence on spin polarization of the ferromagnetic layers.

(3) Nonequilibrium spin injection in ferromagnetic/metal/ferromagnetic structures, and in particular the magnitude of the spin polarization propagated across the ferromagnetic/metal interface.

(4) Study of ultra-thin antiferromagnetic layers via exchange coupling to a ferromagnetic probe layer, and in particular the dependence of the magnitude of the coupling on the atomic-scale structure of the ferromagnetic/antiferromagnetic interface.

(5) Control of magnetic anisotropy in ferromagnetic layers via exchange coupling to a second ferromagnetic or ferrimagnetic layer.

(6) Study of the magnitude of the exchange coupling in ferromagnetically coupled ferromagnetic layers in ferromagnetic/metal/ferromagnetic structures, by determination of the temperature dependence of the magnetization of the ferromagnetic layers.

(7) Fermi-surface driven effects on the magnetic and transport properties of magnetic superlattice structures resulting from the imposed superperiodicity.

Exchange coupled magnetic multi-layered structures form a rich area of research. Progress will most likely be led by experimentation with different material combinations and by attempting to control the microstructure of the interfacial region between the various layers. The ability both to vary the nature of the interface in a controlled manner and to characterize the nature of the interface provides an extremely challenging, perhaps intractable problem. It might well be that the study of extremely small-scale structures, either magnetic dots or 1-D magnetic chains, perhaps grown on terraced substrates, may provide more homogeneous structures with which to examine some of the effects mentioned above.

D. Low-dimensional magnetism

Three prominent issues in surface magnetism concern:

(i) the criteria for and impediments to achieving monolayer magnetism,

(ii) the nature and origin of the surface magnetic anisotropy, and

(iii) the critical behavior of 2-D magnetic phase transitions.

The issues involve the competing influence of electronic and geometric structural considerations. The role of strain fields at surfaces and interfaces in stabilizing perpendicular easy axes of magnetization, relative to the role of the spin-orbit interaction, needs to be assessed. These studies will benefit from high-quality sample preparation and the availability of *in situ*, as well as post-growth, characterization techniques. The importance of growth-induced anisotropies needs to be better appreciated. Test cases of well-characterized model systems need to be established.

The area of critical phenomena in low dimensions provides a particularly satisfying arena for cross-pollination of ideas between experimentalist and theorist. This is because the concept of universality puts the emphasis on characteristic length scales, and not on the details of the interactions or of the structure. The ability of experimentalists to generate data which can be compared to Onsager's 1944 solution of the 2-D Ising model is a long-awaited development that should be close to realization. Issues associated with finite-size effects, inhomogeneities, defects, field-induced-fluctuation phenomena above critical temperatures, etc. all need to be systematically explored in order to make meaningful progress in identifying universal behavior.

Experimental issues associated with the relationship between the measurement probe and the magnetization need elucidation. Invariably surface-sensitive probes of the magnetization couple through ill-defined interaction matrix elements, or are subject to dynamical-scattering effects that introduce intractable corrections whose influence is difficult to assess. The magnetization axis is also subject to reorientation as temperature is raised toward the Curie temperature, especially for vertical easy-axis alignment. The critical region also needs to be defined based on a Ginzburg-Landau criterion in order to determine the temperature range over which data fitting should take place.

E. Excitations

Thermal excitation of spin waves at surfaces have been studied by polarized-electron scattering[143] and sec-

ondary-electron emission.[202] Experiments confirmed theoretical predictions[203,204] that the temperature dependence of the surface magnetization should follow the same $T^{3/2}$ power law as in the bulk. The deviation of the prefactor of the $T^{3/2}$ term from earlier predictions has been explained by assuming an exchange coupling strength of the surface layer to the bulk to be only 30% of the coupling strength between bulk layers.[100] This temperature dependence of the local magnetization at surfaces and interfaces caused by the excitation of spin waves at low temperatures should be further explored as a probe of local exchange interactions in future studies.

Considerable theoretical effort has been directed toward the study of magnetostatic coupling of surface spin waves in multilayer samples. A rich spectrum of coupled excitations, the nature of which depends on whether the ferromagnetic films are aligned parallel or antiparallel to each other, has been predicted.[115,205] Observations of additional modes in Ni/Mo films using Brillouin light scattering supports this picture,[116,206] as also does recent work on Fe/Cr structure.[132] The entire subject of spin excitations in coupled systems is, however, largely unexplored.

Another possible area which has not received sufficient attention is a comparison of results from different techniques. For instance, no comparison of magnon-mode frequencies extracted from FMR and from Brillouin scattering exists.

In multilayer systems the induced periodicity folds the phonon bands into very small Brillouin zones, with a resulting very complex spectrum. Similar effects should be observed in magnetic systems; the resulting multiplicity of superzone gaps must depend sensitively on interlayer exchange coupling. Experimental observation of multilayer magnon bands should be accessible to neutron scattering techniques, especially as more intense sources and larger samples become available.

True surface spin-wave modes (Damon-Eshbach modes) have been observed in Ni/Mo multilayers and on Fe–Cr–Fe sandwiches by light scattering.[116,132] However, light scattering can measure only the $k = 0$ uniform mode and cannot explore dispersion, which would provide information about surface exchange interactions. HREELS may be capable of observing the full surface-magnon dispersion curve, but requires increased energy resolution over that currently available, and better understanding of the scattering processes.

It has been suggested that surfaces order independently from the bulk. If so, it should be possible to explore both the growth of surface coherence and its expansion into the bulk. Quasielastic neutron scattering, perhaps using surface diffraction techniques, is a promising way to pursue such studies. Because truly 2-D systems develop long-range order only in the presence of anisotropy, studies of kinetics of surface magnetism provide information about local surface anisotropy. Kerr effect, magnetometer methods, and spin-polarized scattering may be capable of detecting the kinetics of the ordering process, possibly revealing spin-glass-like surface states or the effects of random local anisotropy.

F. Magnetism and structure

The physical properties of thin films are strongly affected by their structure and composition, to the extent that physical properties can often be predicted if structure and composition are known with sufficient precision. It is not always true that structural and compositional "perfection" are the most desirable traits for magnetic films. Interfacial roughness may, for example, enhance magnetic coupling by breaking wave vector conservation conditions that would otherwise isolate the electronic states of two metals in contact.

In general, it is important to appreciate the spatial scale over which magnetic phenomena occur, and to probe the structural and compositional variations within magnetic materials at the same spatial resolution. Dipolar interactions, domain morphology, and domain walls are "large scale" relative to RKKY and exchange interactions, which are "small scale" and relate to structure at atomic dimensions. Each of these requires attention to structural determination at the appropriate scale.

A variety of techniques for structural analysis is available which permits the determination of atomic-scale structure using diffraction and macroscopic morphology using electron microscopy. For the study of anisotropy phenomena at interfaces and surfaces, roughness may have an important role. Anisotropy, of course, is crucial for determining many physical properties and for stabilizing magnetic ordering in lower-dimensional systems. Defects may provide sources of random anisotropy and exchange that produce spin-glass-like regions. Coupling across nonmagnetic layers also relies on structural aspects of the intervening layers. The presence of pin holes, interdiffusion, and roughness may modify the details of the interactions; but the detection of pin holes and the distinction between roughness and interdiffusion are very difficult to do experimentally. (The reason is that both roughness and interdiffusion appear in a similar fashion in the fitting procedures employed to determine the structure; see Sec. IV, Techniques and Facilities, Diffraction.) The preparation of samples with artificially induced, controlled defects is needed to understand the effect these have on both the structural probes and the physical properties. Examples of these types of studies could be the growth of multilayers on artificially roughened surfaces, studied by x-ray diffraction, TEM, magnetization, and light scattering, with the objective of understanding

whether these defects heal or are enhanced as a function of thickness. These artificially roughened surfaces could be produced by growing samples at different substrate temperatures, depositing small particles on the substrate, using vicinal surfaces, or other procedures.

Generally all types of measurements which correlate atomic and microstructure with magnetic properties are of considerable interest. Microstructural effects such as surface relaxation, surface reconstruction, "roughness", strain (as might be induced by magnetostriction) in thin films, the type and location of defects (including misfit dislocations, threading dislocations, growth ledges, stacking faults), and compositional heterogeneities (segregation, precipitation, impurities) can all have significant influence on the magnetic behavior of thin films, overlayers, and interfaces. These must be carefully monitored with high spatial resolution in order to understand their individual and synergetic effects on local magnetic behavior. Ultimately, the physical structure plays a dominant, perhaps determining role in shaping the magnetic properties.

The mechanical properties of thin films and superlattices have been found to exhibit anomalous behavior, for example the supermodulus effect which is associated with large strains both perpendicular and parallel to the layers. The effect of such mechanical properties on film magnetism has not received much attention.

Strong magnetoelastic interactions provide an important source of coupling among structure, mechanical properties, and magnetism. The strains associated with the layered growth of dissimilar materials in contact with each other restrict the ability of magnetic materials to distort in response to the magnetoelastic energy. Systematic studies are needed to determine the defect structure of such films and multilayers and the changes that occur upon magnetization. The differences that accompany compression and expansion of magnetic films caused by differing lattice parameters is a particularly fruitful direction for future research.

G. Metastability

The work on metastable magnetic structures described in Sec. III (Materials) actually represents the beginning of what should prove to be a fruitful area of research. The initial work on bcc Co and fcc Fe focused only on cubic phases. More recent work has now made it clear that cubic phases are a special case, and that it is more generally expected that body-centered tetragonal phases will be the metastable phases most likely to be stabilized by epitaxial growth. This is true both from a theoretical point of view—where recent calculations[207] found a body-centered-tetragonal metastable phase for Cu—and from an experimental perspective.

Experimentally, the number of possible single-crystal substrate materials is limited. Except for the relatively rare cases of a lattice matched to a cubic phase, the actual growth should in general compensate for the in-plane mismatch by relaxing the interplanar spacing during growth. The resulting tetragonally distorted structure should achieve stability if it is energetically close to a metastable tetragonal phase. The challenge, from the experimental viewpoint, is to characterize carefully the structure of these new tetragonal phases by LEED, EXAFS, or other suitable techniques, and to measure their magnetic properties as well.

H. Anisotropy and magnetostriction

An understanding of anisotropy and magnetostriction in transition-metal materials is of fundamental importance for eventual control and exploitation of these properties in applications. Calculation of these spin-orbit related properties should be attacked both by including spin-orbit coupling self-consistently in electronic structure calculations and by perturbation-theory approaches. The lowered symmetry of surfaces and thin films means that the dominant terms will generally be second order in the spin-orbit coupling, and thus larger than in most bulk materials. The important issues are whether the Fermi surface can be calculated with sufficient accuracy to yield meaningful results and whether the precision of the energy calculations can be improved to the level of spin-orbit energies.

Ab initio calculations of crystal fields and crystal-field parameters have not yet reached the level of accuracy that is required for most purposes. Progress in that direction is needed. Opportunities for theoretical research in the area abound.

From an experimental perspective, the issues of understanding how interface anisotropy and magnetostriction can be manipulated and/or controlled will determine whether future devices can be developed. For example, what types of materials can be used at the interface of a compositionally modulated layer to cause or eliminate perpendicular or in-plane anisotropy? What can be done to make significantly softer magnetic films (NiFe, Co)? Can surface anisotropy be used to compensate bulk anisotropy in thin films? Can the dispersion in anisotropy be reduced by growing more perfect films? These very practical questions require a more thorough understanding, both theoretical and experimental, of many materials.

I. Magnetoresistivity

Galvanomagnetic effects in very pure materials have been extensively used[27] to determine very subtle properties of their electronic structure, in particular the topology of the Fermi surface in metals. The effects, on the other hand, are more difficult to understand and interpret when macroscopic spatial heterogeneities play a

fundamental role. Such is the case with extended defects, shape and size imperfections, and surface roughness.

As mentioned previously (see Sec. II, Theoretical Background, Transport Properties in Magnetic Systems), positive MR—an increase in the resistance upon application of a magnetic field—is associated with the convoluted character of crystal electron orbits in a magnetic field. Negative MR—a decrease in resistance when the field is applied—implies increased order, a reduction in the strength of the electron scattering or a suppression of its sources. Negative MR in ferromagnetic materials is qualitatively understood, and is caused either by the removal of domain walls upon application of the field or by the introduction of a gap in the spectrum of the spin waves, which are then less effective in scattering the conduction electrons.

The recently discovered "giant" negative magnetoresistance in (001)Fe/(001)Cr superlattices[96] can be as large as a factor of two, and has been attributed to the spin-dependent transmission of conduction electrons between Fe layers through the Cr interlayers. This explanation is obviously only qualitative. The phenomenon deserves considerable attention: (1) its fundamental origins remain unclear, (2) it may perhaps be found in systems more general than the specific one in which it was discovered, (3) it is most probably related to the antiparallel coupling of the nearest neighbor Fe (individually ferromagnetic) layers, and (4) it will almost certainly have important technological applications (see Sec. V, Applications, A. 2: Exchange Biasing of Magnetoresistive Heads).

J. Micromagnetics

The configuration of domains and domain walls is strongly affected by the presence of surfaces or interfaces at which there are demagnetizing fields and large contributions to the magnetostatic energy. Recent SEMPA investigations[34] have provided detailed insight into how a Bloch wall in the bulk of a material like Fe or permalloy turns over into a Néel wall at the surface. The Bloch wall can turn over into the Néel in two different directions such that the walls are offset. Micromagnetic calculations employing a continuum approximation show quantitative agreement with experimental asymmetric surface Néel wall profiles and offsets. When two offset walls meet there is a topological singularity in the magnetization. The size of the core of such a singularity remains a challenge to measure with yet higher resolution SEMPA. A number of singularities have been discussed theoretically[208,209] and invite investigation with SEMPA. Other questions as to how the magnetic microstructure changes as the specimen thickness is decreased or what is the magnetic structure of one ferromagnetic film on top of another present further research opportunities.

A very interesting question is whether a film of a single monolayer (if such could be realized in practice) would support domains. Recent work[210] suggests that although uniform magnetization is expected for in-plane magnetization, domains are energetically favorable for perpendicular magnetization when the perpendicular surface anisotropy exceeds a critical value. It would be of great interest to know how the size of the domains and walls could be expected to vary in films of two, three, or more layers. It may be possible with SEMPA to investigate such domains and walls if suitable films with appropriate perpendicular anisotropy can be prepared. Further, an extension of the theory to finite temperatures would allow direct comparison with experiment. In general, micromagnetic theory should be extended to the very small dimensions of these systems so that calculation of domains and domain walls would be possible. This would help clarify the relationship between surface hysteresis loops measured with electron spectroscopies, and the bulk hysteresis curves characteristic of a deeper region.

At the microscopic level, domain walls may be zigzag instead of straight and sharp. If the domain wall is the transition between two written bits on a recording medium, the zigzag wall gives rise to noise and represents an ultimate limitation on the recording density. Noise is also affected by the correlation of magnetization reversals for individual grains; this property may be examined by experiments such as the anomalous Hall effect.[211] (These sources of noise may be reduced by decreasing the exchange coupling between grains.) With the push to ever higher density of information storage, there is much research needed to understand and control magnetic microstructure at the microscopic level.

At the boundaries of a ferromagnet small closure domains form to reduce the energy further. It is often desirable to control the closure domains. This can be done by varying the shape of a magnetic element. Further, with a thin film, closure domains can be reduced by providing a return path for the edge flux through a second layer. For example, a thin-film recording head might consist of two magnetic layers separated by a nonmagnetic layer of sufficient thickness to avoid exchange coupling of the layers. Further development of means to control the magnetic microstructure is facilitated by new means of obtaining high-resolution images of the magnetization.

Domain nucleation is an issue which, if it were understood, could provide considerable insight into both explaining experimental observations and designing new materials. Unfortunately, domain nucleation in real materials probably involves a complicated relationship among the electronic structure near a defect, thermal fluctuations of the neighboring spins, and the normal macroscopic forces of micromagnetics. An un-

usual opportunity to study this effect may be found in certain barium-ferrite particles which simultaneously display a high degree of crystalline perfection, a smaller coercivity than might be expected, a larger time dependence of coercivity than expected, and a thermally activated magnetic dead layer on the surface.[212,213] A reasonable conjecture is that the latter three properties are related and that fluctuations in the surface dead layer are leading to domain nucleation and switching. Experimentally, a detailed characterization of the surface crystal structure might immediately lead to candidate nucleation sites. It might, alternatively, provide a foundation for a detailed theoretical treatment which could include the local fluctuations balanced against the micromagnetic forces. It is worthwhile noting that an alternative, although somewhat less easily characterized system, is the γ–Fe_2O_3 particle, where it is possible that Co surface doping removes nucleation sites.[214]

A predictive understanding of hysteresis loops, of which the coercivity and remanent coercivity form particularly interesting features, would be obviously beneficial. In some materials the coercivity and nucleation field will coincide, but in many other cases nucleation will not be the fundamental barrier. A fruitful approach for some of these latter materials should be an extension of a currently available domain-wall pinning theory[31] toward greater quantitative accuracy. Calculations of moment, exchange, and interface anisotropies at grain boundaries will be particularly useful. Careful experimental study of select systems should provide the nature of the grain boundary, and studies of time dependence may help determine the shape of the energy barrier. In other materials, such as those exhibiting sharp, well-defined grains, further implementation of the approach of Ref. 32 would be appropriate. Here the goal would be to compare the results of accurate implementation of micromagnetic theory including, if necessary, domain nucleation, to equally accurate experiments for a variety of systems beyond the CoNi thin films originally treated. This will help determine the physical limits of the theory.

In general, future advances will substantially depend on careful comparison among atomic structure, micromagnetic features observed by high spatial resolution techniques, and accurate computer simulations. Investigations will consider progressively smaller spatial scales.

K. Magnetics technology

Because of the many applications of magnetic materials and phenomena, new insights into the basic physics of magnetism often have technological implications. An investigation of the fundamental properties of the interface, whether it be the surface-vacuum interface, the interface between thin film and substrate, or the interface between magnetic layers, also provides an opportunity to contribute to the solution of many technological problems. A deep understanding of the interactions at interfaces will allow scientists to control material properties. For example, a suitable underlayer for a recording medium can control grain size and orientation. A fundamental understanding of interface decoupling layers may, along with the ability to control segregation to grain interfaces, lead to reduced noise in magnetic recording media or increased coercivity in permanent-magnet materials. The current understanding of interface-induced anisotropy or of exchange biasing at an interface is insufficient for engineering purposes. Clearly, there are many research opportunities into fundamental questions which also represent research opportunities in magnetics technology.

L. Conclusion

An effort has been made to describe briefly the current status of research in surface, interface, and thin film magnetism and highlight some of the issues and research opportunities. Even though the discussion was necessarily brief, the report is lengthy owing to the diversity of the field and the high and increasing level of research activity. The assembled panel collectively has a wide background in the subject area; nevertheless, it is impossible to be absolutely comprehensive in coverage. Still other issues which offer interesting research opportunities could have been discussed.

The research opportunities in surface, interface, and thin film magnetism are exciting and many. Advances in the growth and preparation of magnetic systems, an active research area itself, have led to materials with different crystal phases, altered lattice constants, layered structures, and so on, in short, to new magnetic systems. Characterization of these new materials has caused refinement of existing techniques and development of new ones to determine structural, electronic, and magnetic properties. The theory of magnetism in these lower dimensional systems, often aided by the availability of powerful computational facilities, has been important in stimulating and understanding experimental work. These factors jointly create especially significant opportunities for research in this area.

Magnetism in bulk solids is a well-developed research area which has provided a fertile testing ground for quantum mechanics, theories of many-body interactions and collective phenomena, and critical phenomena. Surfaces, interfaces, and thin films represent new magnetic systems and are, further, building blocks for more complex systems such as multilayers. With the help of new materials technologies, these systems can be prepared in metastable phases which have no bulk counterpart. Size effects and lower dimensionality add interesting new facets to the study of magnetic proper-

ties of surfaces, interfaces, and thin films. In short, we have new materials, exhibiting new properties, and presenting many fascinating fundamental questions to be answered.

Many of the developments in the study of magnetism have been driven by requirements of magnetics technology. This multibillion-dollar-a-year industry spans technologies from magnetic media for information storage to permanent magnets for motors. Creating a new materials system and understanding its magnetic properties has the potential to make a significant contribution to technology. What may be fundamental research questions about interactions at interfaces may ultimately provide the information to control knowledgeably the coercivity and anisotropy in a thin film or the exchange coupling between grains with a consequent impact on information storage devices. One of the exceptional aspects of magnetics research is that progress in fundamental issues and the solving of technological problems often go hand in hand.

The research opportunities in surface, interface, and thin film magnetism touched on in this report range from basic to applied issues and include both experimental and theoretical questions which should challenge researchers in university, government, and industrial laboratories for a number of years. It is an exciting area to work in: there is much to be done.

Table of acronyms.

ALS	Advanced Light Source, Lawrence Berkeley Laboratory
APS	Advanced Photon Source, Argonne National Laboratory
CVD	Chemical Vapor Deposition
DPC	Differential Phase Contrast
ECS	Electron Capture Spectroscopy
EXAFS	Extended X-ray Absorption Fine Structure
FEL	Free-Electron Laser
HEED	High-Energy Electron Diffraction
LEED	Low-Energy Electron Diffraction
MBE	Molecular Beam Epitaxy
MOCVD	Metal-Organic Chemical Vapor Deposition
MOKE	Magneto-Optic Kerr Effect
MR	Magneto-Resistive or MagnetoResistance
MFM	Magnetic Force Microscopy
NSLS	National Synchrotron Light Source, Brookhaven National Laboratory
PNR	Polarized Neutron Reflectometry
RE	Rare Earth
RHEED	Reflection High-Energy Electron Diffraction
RKKY	Ruderman-Kittel-Kasuya-Yosida interaction
SEM	Scanning Electron Microscopy
SEMPA	Scanning Electron Microscope with Polarization Analysis
SHG	Second-Harmonic Generation
SMOKE	Surface Magneto-Optic Kerr Effect
SPEELS	Spin-Polarized Electron Energy Loss Spectroscopy
SPLEED	Spin-Polarized Low-Energy Electron Diffraction
SPSEE	Spin-Polarized Secondary-Electron Emission
SQUID	Superconducting Quantum Interference Device
SR	Synchrotron Radiation
STEM	Scanning Transmission Electron Microscopy
STM	Scanning Tunneling Microscopy
TEM	Transmission Electron Microscopy
TM	Transition Metal
UPS	Ultraviolet Photoemission Spectroscopy
VSM	Vibrating Sample Magnetometer
VUV	Vacuum UltraViolet
XPS	X-Ray Photoemission Spectroscopy
1-D	One-Dimensional
2-D	Two-Dimensional
3-D	Three-Dimensional

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