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L.M. Falicov

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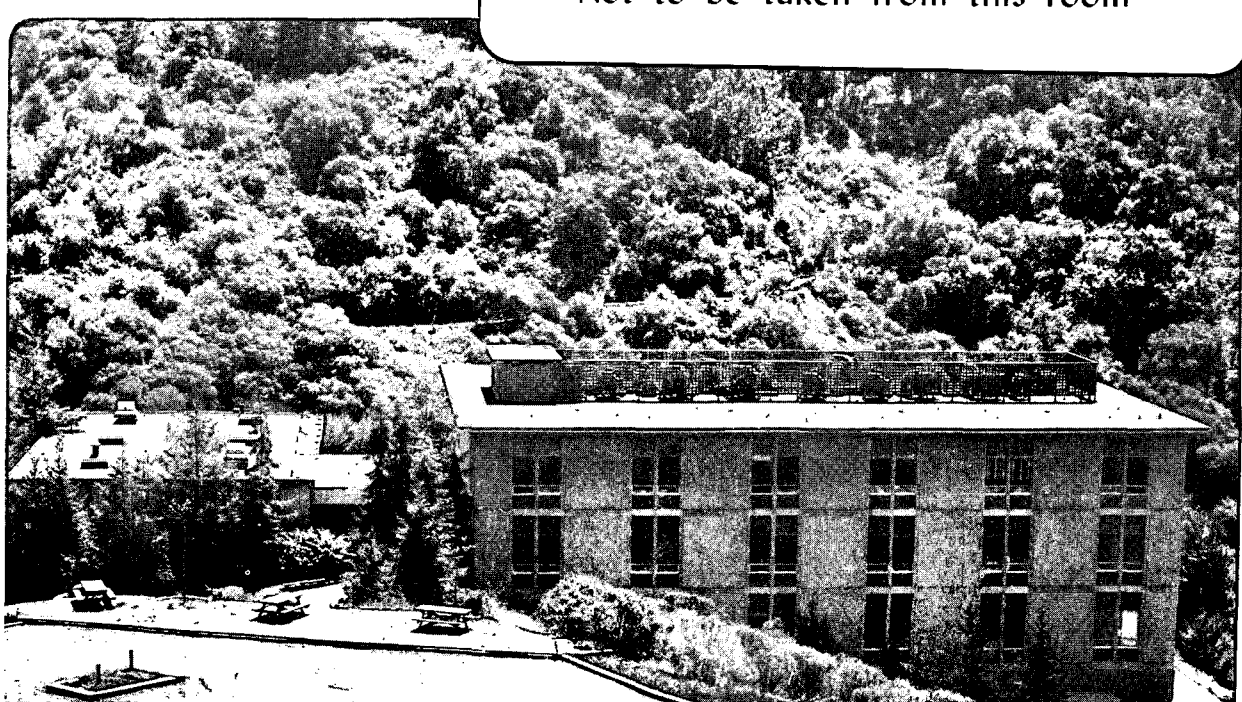

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SURFACE, INTERFACE AND THIN-FILM MAGNETISM: AN OVERVIEW*

L. M. Falicov

Department of Physics
University of California
Berkeley, CA 94720

and

Materials Sciences Division
Lawrence Berkeley Laboratory
Berkeley, CA 94720

April 1991

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L. M. FALICOV
Department of Physics, University of California, Berkeley CA 94720, and
Materials Science Division, Lawrence Berkeley Laboratory, Berkeley CA 94720.

ABSTRACT

A brief review of the state of the art in the field of surface, interface and thin-film magnetism is presented.

INTRODUCTION.

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 existence of the electron spin and in the Pauli exclusion principle. It leads 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. In particular, 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 [1].

Several technical developments are responsible for the intense activity in this field. In addition to application-driven pressures, 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 produced, 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 very 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.

OVERVIEW: THEORY.

<u>Electronic calculations</u>. The prediction of magnetic structures, in

bulk materials as well as in surfaces and interfaces, is still an imperfect science, even though enormous progress has been achieved [2]. Complex structures have been predicted for bulk, surface and composite structures. Many times such calculations agree with experimental results, but there are notable exceptions.

Calculated Fermi surfaces of magnetic metals show good agreement with experiment [3] in some cases (Fe), not as good [4-6] for others (Co, Ni). Reliable Fermi surfaces however are necessary for predicting as well as interpreting and understanding 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 are, in general, quite reliable [7].

Critical phenomena. There are, in addition, fascinating surface effects related to a variety of critical phenomena: behavior and transitions involving the decay in short-range order [8], the interplay between surface and bulk effects [9] (including the persistence of order on surfaces 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 non-universal behavior of magnetic overlayer systems when the coverage is fractional [10].

Transport properties. 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, 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. Increases in resistance of many orders of magnitude are observed in particularly pure, single crystals at very low 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 [11] 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 two facts: (1) the spin-up and the spin-down electrons have different band structures and different phase space available for transport and for scattering; (2) the external field changes the domain structure, and produces a single-domain crystal. Under those conditions three effects take place. The electrons with different spin encounter different spatial arrangements which change with applied field; the electron trajectories, because of the presence of a now uniform internal field, become less convoluted; and the removal of Bloch walls eliminates a source of electron scattering [12]. All three effects result in longer mean-free paths upon application of a magnetic field, i.e. a negative magnetoresistance. Similar negative magnetoresistance effects have been found in multilayer systems, as

described below.

Micromagnetics. 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 [13]. Input to the calculations includes exchange parameters, crystalline anisotropy constants, and sample microstructure. 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 requires that the motion of the magnetization be traced in time to ensure accuracy. Various calculations, semi-quantitative in nature, have provided considerable insight. Quantitative accuracy is normally prohibitive.

OVERVIEW: SYSTEMS.

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 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 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.

Transition-metal substrates 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 [14]. The in-plane expansion leads to an interplanar contraction and a strong Fe-Ru band hybridization 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 [15]. It is interesting to note that these trends are mirrored in the behavior of dilute Fe alloys in 4d-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.

Surface perfection manifests itself in the quest to verify the theoretical predictions regarding possible ferromagnetism [16-18] 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 bulk 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. 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 issue. It is expected that increased experimental resolution in imaging and an enhanced ability to create ultraflat surfaces will be forthcoming and will help resolve these problems.

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 either 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) is 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 referred to as metastable structures.

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 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 arrangement. As an example, 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) [19]. Both faces are predicted by theoretical calculations [20]. This indicates that fine details of total energy calculations may be manifest in metastable thin films.

Another example of a metastable phase is given by the 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 [21] was successfully formed by epitaxial growth on GaAs. Total-energy calculations [20] yield the bcc-Co phase with the observed lattice constant, and correctly predict it to be ferromagnetic.

There is also a theoretical indication that there should be bcc phases of Ni, both ferromagnetic and nonmagnetic, even though in nature Ni only appears 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 the strong influence of the ferromagnetic substrate made magnetic characterization difficult.

Semiconducting 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 x 2) reconstruction of the metal films upon these substrates. Although bcc Co has been successfully grown on GaAs [21] and bcc Fe on Ge, GaAs and ZnSe [22], there are important unresolved issues of interface chemistry with these systems.

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. Generally it has been found that the growth of rare earths can be accomplished quite conveniently on a transition metal [23-25]. 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.

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 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 by chemical vapor deposition [26]; titanium oxides were grown on sapphire by MBE [27]. 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 $REFeO_3$ (where RE is a rare earth) [28].

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 or MBE, although recently titanium-oxide-titanium superlattices have been grown by the chemical vapor deposition techniques.

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 not yet available.

OVERVIEW: PHYSICAL EFFECTS.

Proximity effects. In some systems, interface effects of a purely magnetic origin extend beyond the interface and into the bulk, thus giving rise to proximity effects. 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 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 [29-31]:

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 sizeable 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 [32,33], nickel tends to be marginally more magnetic at the surface [34], and it is possible that some crystallographic faces of vanadium exhibit a magnetic moment [29].

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 [35], the enhanced moment of the chromium surface tends to propagate over several layers into the bulk [36], chromium acquires a large moment in the proximity of iron [32] and/or cobalt, and vanadium and palladium may develop sizeable magnetic moments in the proximity of iron and/or cobalt.

Exchange coupling across interfaces. Magnetic exchange coupling between ferromagnetic and antiferromagnetic layers was discovered and studied in various systems, e.g., the Co/CoO and $Ni_{81}Fe_{19}/Fe_xMn_{1-x}$ systems. In general there is a large discrepancy (of approximately two to three orders of magnitude) between theoretical estimates of the interfacial exchange coupling energy and the measured values. Although various models have been proposed to account for the large discrepancy between experiment and theory, none is yet fully accepted.

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 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 [37].

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.

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 crsytals. Examples include single-crystal bcc Ni/Fe bilayers, 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 posssible application in a variety of magnetic recording devices.

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 (Ruderman-Kittel-Kasuya-Yosida or RKKY interaction). Early work in rare-earth multilayers [38,39] 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. 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 [40]. Rare-earth sheets are 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.

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 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 reference [40]. Other evidence for spin-dependent transmission has been seen in magnetotransport experiments in the Fe/Cr/Fe system.

Coupling through nonmagnetic layers. Great interest has developed in the last two years in the magnetic and magneto-transport properties of layers of ferromagnetic TM -- Fe, Co, Ni and the $Ni_{80}Co_{20}$ alloy in particular -- separated by layers of variable thickness of ordinary transition or noble metals metals (Cr, Ru, Re and Cu). Two related effects are found: (1) successive ferromagnetic layers couple variously in either antiferromagnetic and ferromagnetic arrangements, depending on the thickness and nature of the intervening non-ferrmagnetic metal; and (2) samples with antiferromagnetic coupling between successive layers exhibit either a small (e.g. Co/Ru) or a giant (Fe/Cr) negative magnetoresistance. These effects have been observed by a variety of techniques: spin-polarized electron-scattering [41], magneto-optic-Kerr-effect and Brillouin scattering studies [42, 43], galvanomagnetic measurements [44,45], magnetization studies [45,46], neutron diffraction [46] and ferromagnetic resonance [43] among others [47]. Various theoretical models have been proposed to account for these phenomena [48-54], although the coupling mechanism is, at present, not yet fully understood. It is apparently too large a coupling to be accounted for by magnetostatic effects, and although the coupling strength oscillates with normal-metal-film thickness, the period of the oscillation is much too large to be caused by RKKY, unless it is dominated by very small pieces of the normal-metal Fermi surface. The giant magnetoresistance found in the Fe/Cr multilayer samples is of great interest for potential recording head applications.

There have been numerous other 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 [55,56], such as Fe/Ag. Exchange coupling of successive Fe layers in this system has been inferred [56] from the temperature dependence of the magnetization at low temperatures. A calculation [57,58] 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 fairly recent elegant experiments [59] 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 [60]. It has been proposed [61] that tunneling between two ferromagnets could depend on the relative alignment of the magnetization of the two ferromagnetic layers; this effect was subsequently observed [62] 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 [63] in ferromagnet/insulator/superconductor junctions. More recently, similar effects have been observed in [64] Ni/NiO/Co tunnel junctions.

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 observed in Dy superlattices [65] and films [66], and 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 [67]. 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.

Superlattice effects. Many of the effects described above can be conveniently studied in simple snadwiches or in multilayered films since the latter consist of a superposition of single films [68]. 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. The drawback is, of course, that by its very nature any single, bi- or tri-layered film effect can only be obtained in a statistical sense, averaged over many 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. 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 [69]. Whether this scattering is sufficient to break down the existence of extended states perpendicular to the layers and in effect confines the electrons to individual layers is not clear.

A superlattice effect which does not require perfection at the atomic level is the development of magnon bands in ferromagnet/normal-metal superlattices [70]. 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.

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LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720