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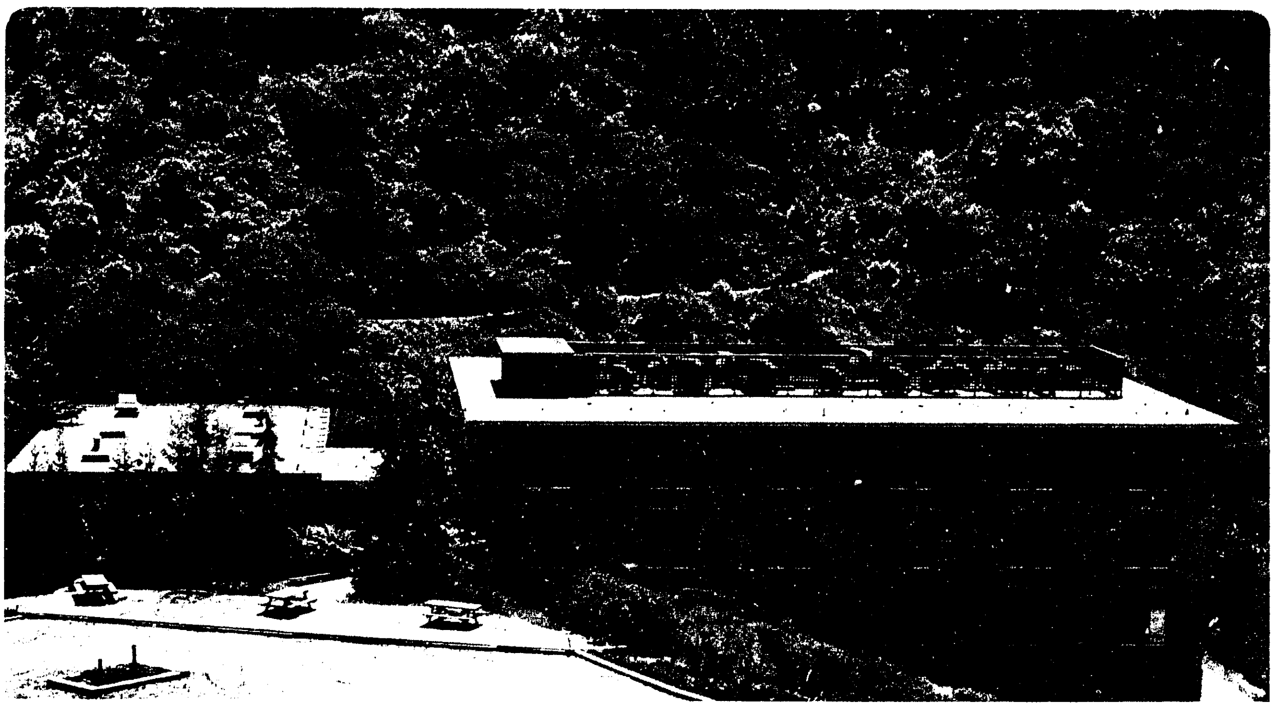

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SURFACE, INTERFACE AND THIN-FILM MAGNETISM: AN OVERVIEW

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INTRODUCTION

In the last quarter of the 20th century, 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 has been 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 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 which provide a wealth of properties. From these features useful engineering and technical applications arise [1]. Several developments are responsible for the intense activity. Three major technical advances are to be noted:

(1) The advent of new sample-preparation techniques which 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.

Electronic calculations. 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. 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 electronic structure and Fermi surfaces of magnetic metals show good agreement with experiment in some cases (Fe), not as good 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 "phenomenological" methods are constructed to reproduce known experimental or ab initio results, predictions are, in general, quite reliable.

Critical phenomena. There are fascinating surface effects related to a variety of critical phenomena: behavior and transitions involving the decay in short-range order, the interplay between surface and bulk effects (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.

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, has been extensively employed to determine electronic structures, Fermi surfaces in particular. In ferromagnetic systems, which in the absence of an applied field consist of several magnetic domains, the phenomenon of negative magnetoresistance 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 in a single domain become less convoluted; and the removal of Bloch walls eliminates a source of electron scattering. 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.

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. It is a classical (i.e. non quantum-mechanical) many-body problem with great computational expense. The memory dependence of the problem requires that the motion of the magnetization be traced in time.

OVERVIEW: SYSTEMS.

Surface and monolayer films. 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.

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 at the interface to drive the film into other, metastable crystallographic structures.

Semiconducting substrates. Single-crystal semiconductor substrates provide a very attractive template for the epitaxial growth of metal films.

Rare earths. The growth of rare earths provides a particularly fertile ground for the study of magnetic phenomena in thin films and multilayers. 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.

Oxides. An important system which has not been studied extensively is the growth of epitaxial oxides. Oxides exhibit interesting magnetic properties -- such as antiferromagnetism -- and are the basis for a variety of devices, especially when used in conjunction with ferromagnets.

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 also chemical vapor deposition techniques have been used.

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. In particular:

1.- The magnetic moments of Co and Ni are virtually saturated.

2.- The magnetic moment of Fe, 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, V and Pd, may acquire a sizeable magnetic moment in the proper environment.

5.- Free surfaces tend to increase the magnetic moment of an element; the surface of Cr has a much larger moment than the bulk, Ni tends to be marginally more magnetic at the surface, and it is possible that some crystallographic faces of V exhibit a magnetic moment.

6.- Proximity of a nonmagnetic metal tends to suppress the magnetic moment of some magnetic elements.

7.- The proximity of a strongly magnetic element tends to induce or enhance magnetic moments on the neighboring, susceptible elements. Thus Fe becomes more magnetic in the proximity of Co, the enhanced moment of the Cr surface tends to propagate over several layers into the bulk, Cr acquires a large moment in the proximity of Fe and/or Co, and V and Pd may develop sizeable magnetic moments in the proximity of Fe and/or Co.

Exchange coupling across interfaces. Magnetic exchange coupling between ferromagnetic and antiferromagnetic layers was discovered and studied in various systems, e.g., Co/CoO and $Ni_{81}Fe_{19}/Fe_xMn_{1-x}$. In general there is a large discrepancy (about 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.

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). Evidence for spin-dependent RKKY transmission has also been seen in magnetotransport experiments in the Fe/Cr/Fe system.

Coupling through nonmagnetic layers. Great interest has developed in the last three years in the magnetic and magneto-transport properties of layers of ferromagnetic metals -- 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 (Ti, V, Cr, Cu, Zr, Nb, Mo, Ru, Rh, Pd, Ag, Hf, Ta, W, Re, Ir, Pt and Au). 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 (e.g. Fe/Cr) negative magnetoresistance. These effects have been observed by a variety of techniques: spin-polarized electron scattering, magneto-optic- Kerr-effect and Brillouin scattering studies, galvanomagnetic measurements, magnetization studies, neutron diffraction and ferromagnetic resonance among others. Various theoretical models have been proposed to account for these phenomena, although the coupling mechanism is, at present, not yet fully understood. It is too large a coupling to be accounted for by magnetostatic effects. Although the coupling strength oscillates with normal-metal-film thickness, the period of the oscillation varies as a function of the structure (smoothness) of the interface. For ordinary interfaces it is much too large -- about ten lattice spacings -- to be caused by RKKY; for very carefully manufactured interfaces the period is much smaller, and consistent with the RKKY interaction, i.e. approximately two lattice spacings for Cr, in agreement with the known antiferromagnetism and the Fermi surface. The giant magnetoresistance found in the Fe/Cr multilayer samples is of great interest for potential recording head applications.

Tunneling between a spin-polarized superconducting film and a coupled ferromagnetic layer has been extensively used to study the magnetic properties of thin ferromagnetic layers.

Tunneling between two ferromagnets depends on the relative alignment of the magnetization of the two layers; this effect was observed in Fe-Ge-Co and in 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.

Superlattice effects. Many of the effects described above can be conveniently studied in simple sandwiches or in multilayered films. Multilayers provide the possibility of ex-situ studies without 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 a class of effects which cannot be observed in a small number of layers because they rely on the periodic nature of the multilayer. These are the 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. 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.

PROSPECTS, OPPORTUNITIES AND FUTURE DEVELOPMENTS

Theory. As resources and computer capabilities increase several important calculations will become feasible: (1) better understanding of the solid-vacuum interface; (2) magnetic properties of interfaces between two different materials with different symmetries, e.g., Fe/Cr/Fe sandwiches and Fe/Cr superlattices; (3) inclusion of structural relaxation at surfaces and interfaces; (4) prediction of magnetic structures by means of *ab initio* total-energy calculations; (5) theoretical determination of secondary magnetic properties, such as anisotropy and magnetostriction.

Magnetic Moments at Surfaces and Interfaces. This general area is still only sketchily explored and is a rich ground for basic research with possibly many practical applications.

Magnetic Coupling at Interfaces. One of the most exciting areas of both current and future research. Materials engineering of these magnetic systems allows for the optimization and control of such basic magnetic properties as saturation magnetization, anisotropy, coercivity, and magnetic domain structure. It seems clear that these types of structure will be of increasing importance in the magnetic-recording industry.

Low-Dimensional Magnetism. Three prominent issues in surface magnetism concern: (1) the criteria for and impediments to achieving monolayer magnetism; (2) the nature and origin of the surface magnetic anisotropy; (3) the critical behavior of 2-D magnetic phase transitions.

Excitations. Thermal and electromagnetic excitation of spin waves at surfaces.

Magnetism and Structure. Their interdependence in thin films and multilayers. Magnetoelastic interactions are paramount here.

Metastability. The discovery of new structural and magnetic states of well known and new structures, with potential applications.

Anisotropy and Magnetostriction. The understanding of anisotropy and magnetostriction in transition-metal materials is of fundamental importance for eventual control and exploitation of these properties in applications. Ab initio calculations of crystal fields and crystal-field parameters have not yet reached the level of accuracy that is required for most purposes.

Microstructure. An understanding of the relationship between magnetism at surfaces and interfaces, and the microstructure is important. 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.

Magnetoresistivity. The "giant" negative magnetoresistance in (001)Fe/(001)Cr and similar superlattices requires further study. It will certainly have important technological applications.

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 Scanning Electron Microscopy with Polarization Analysis studies have provided detailed insight into how domain walls in the bulk of a material behave when they reach the surface. It is of great interest to know how the size of the domains and walls vary in films of two, three, or more layers.

Domain nucleation is an issue which, if understood, could provide considerable insight into both explaining experimental observations and designing new materials.

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.

Summary. 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.

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