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MAGNETISM OF THIN FILM MULTILAYERS: AN ANALOGUE OF INTERACTING PLATELETS

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Magnetism of thin film multilayers: an analogue of interacting platelets.

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ABSTRACT. Progress is being made toward manufacturing materials with magnetic properties tailored to the desired application. This result is reached in several steps, which are monitored with different optical techniques such as polarized neutron reflectometry. First, ferromagnetic, metallic films (of Fe, Co, Ni, Gd), a few nanometers thick, are prepared by vapor deposition. Their magnetization can be tuned by changing the chemistry or thickness of the films, and can be biased by embedding the films into a matrix of antiferromagnetic material. Ensembles of metallic films (multilayers or superlattices) can be created, with a magnetic coupling between adjacent layers regulated by the nature and thickness of the spacer. For increasing spacer thickness, the alignment of neighboring magnetic layers switches between a parallel (F) and an opposite arrangement (AF) in an oscillatory manner. In multilayer structures possessing more than one kind of magnetic atom complex magnetic phase diagrams have been predicted to occur, with properties that are strongly influenced by the presence of a surface. With these characteristics, the phenomenology of magnetic multilayers draws a close similarity to the physics of interacting platelets.

1. Introduction

The role of this NATO Advanced Scientific Institute is to show how to manufacture materials with tailored physical (mechanical or electric) properties, by intervening at the mesoscopic -rather than the microscopic- level. At the atomic scale, nature provides a few fixed entities that cannot be easily modified. Instead macromolecules, bubbles and plateletes can be manufactured out of some chemical substance with sizes that range from a few nanometers to some microns. If these particles are diluted in a medium, the particle-medium interaction can be changed almost at will. By increasing the particle concentration, controlled assemblies are generated with phase diagrams which are novel and, to a certain extent, predictable. This is hardly surprising: while the interaction between atoms is rigidly defined by their nature, the effective potentials between macromolecules, bubbles or platelets can be altered by changing the geometry of the constituents, the relative distance or their surface chemistry. The objective of this communication is to show that a parallel development is taking place in regard to magnetism, providing new hope of engineering materials having predictable magnetic properties.

Magnetism in solids is a well researched subject[1,2] and it would be ludicrous to

attempt to review it in a few pages. Suffices to say that, for certain materials, a certain temperature exists below which a time-invariant magnetic order is present. The magnetic structure can be represented by assigning permanent magnetic moments (which can be drawn as tiny arrows) to the magnetic atoms. The moment arrangement can yield a net magnetization, in ferri- and ferromagnets, or (when the magnetic moments compensate each other exactly) to zero magnetization (antiferromagnets). Fig.1 illustrates some of the possible magnetic arrangements for the unit cell of a simple cubic lattice[3]. Fig.1a shows a ferromagnetic arrangement. Fig.1b and c show two of the possible antiferromagnetic arrangements, which differ for the orientation of the magnetic moments with respect to the axis of propagation of the antiferromagnetic spin density wave (parallel to it in Fig.1b, perpendicular in Fig.1c). As crystalline structure is created by filling the space with symmetry related points, a magnetic structure is created by filling the space with arrows, each indicating direction and size of the magnetic moment of an equivalent atom. It is not surprising that for 230 crystalline space groups the number of magnetic space groups amounts[4] to 1421.

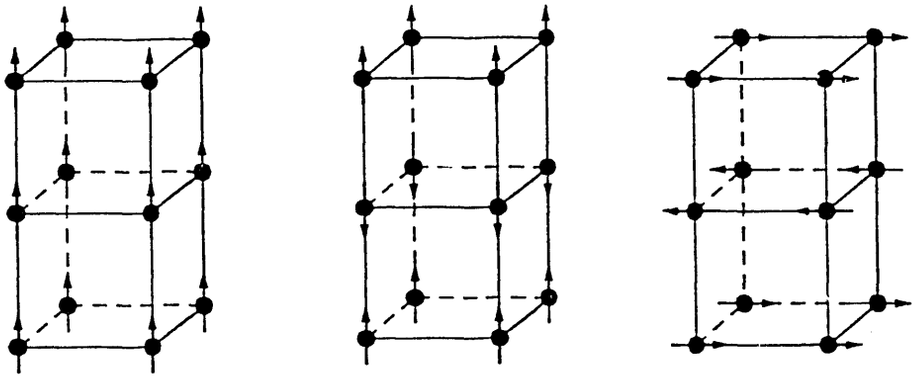

Fig.1 A simple cubic lattice and three possible types of magnetic order: a) ferromagnet; b) and c) two antiferromagnetic arrangements.

2. Optical probes

Surface, interface and thin film magnetism has been reviewed recently in a plenary paper[5], which surveys also the techniques recently devised to study such magnetism. Thus only a short mention will be given here of those techniques, treating more extensively only polarized neutron reflectivity (PNR), because we will use repeatedly PNR experiments to illustrate the physics of our subject.

The most traditional measurement of magnetization is magnetometry. Pushed to a sensitivity of 10^{-8} emu with the advent of the Superconducting Quantum-Interference Device (SQUID), a commercial unit is now capable of measuring the magnetization of a single atomic layer of Fe with a 1 cm^2 surface. However, the magnetometer measures at the same time the magnetization of substrate and sample holder whose contribution, to be subtracted, may dominate the signal. A host of more selective techniques have been developed to complement conventional magnetometry. For instance, ferromagnetic resonance can determine the magnetization of thin films, and to explore it in more detail. Angle-resolved magnetic resonance has been used[5] to determine magnetic anisotropy, and

the width of the resonance line has provided information on the uniformity of the magnetization along the layer thickness. Various microscopic technique have enabled to select chosen areas of the sample. For the magnetooptic Kerr effect suffices a size of the illuminated spot of 10μm diameter (in the Kerr effect, what is measured is the rotation of the polarization vector of a laser beam after reflection from a magnetized surface). Even smaller areas are sampled by scanning electron microscopy with polarization analysis (SEMPA). This technique measures the spin polarization of the secondary electrons which form the scanning electron microscope topographic image: the image of the magnetic domains is obtained with a resolution as high as 10 nm. Perhaps the ultimate resolution is attained by Mœssbauer effect of certain nuclei (like Fe^{57}) implanted on very selected sites of the sample. By Mœssbauer effect, the magnetic field at the resonant nucleus is measured, and from this the local magnetization (in size and direction) is inferred.

A great deal of information on magnetic thin films is obtained also by diffraction of different "lights" such as spin-polarized low-energy electrons, X-rays and neutrons - in short, diffraction of almost any particle that has a magnetic moment, and a wavelength comparable to the thicknesses to be measured in the film. Spin-polarized low energy electron diffraction is widely used to study surface magnetic states, since 10-100 ev electrons can only penetrate a few Ångstroms. In contrast, X-rays and neutrons can detect magnetic moments deep in the material as well as at the surface. Neutrons have a sizeable magnetic moment: the interaction with the magnetic electrons of an atom like iron is comparable to the interaction between the neutron and the nucleus of that atom. As a consequence neutron reflection experiments are well suited to study the detailed magnetic configurations of magnetic materials, even in the form of thin films.

In analogy to the propagation of electromagnetic radiation (including X-rays) through matter, a refractive index for neutrons can be defined as[6]

$$n = k / k_0 = (1 - V / E)^{1/2} = [1 - 4\pi(N \pm M) / k_0^2]^{1/2} \qquad (1)$$

where k_0 and k and are respectively the neutron wavevectors in vacuum and in a medium. V is the potential energy of the medium, E is the neutron kinetic energy, N is the nuclear scattering density and M is the magnetization. The signs in front of M depend on the direction of the neutron moments with respect to the magnetization (parallel: + ; antiparallel: -): a ferromagnetic material is birefringent.

The specular reflectivity for a neutron wave incident on a flat surface is a function of wavevector transfer $\mathbf{Q} = |\mathbf{k}_{0f} - \mathbf{k}_{0i}| = 2k_0 \sin\theta$, where θ is the angle of incidence with the surface (Fig.2), and $\mathbf{k}_{0i}$, $\mathbf{k}_{0f}$ are the initial and final wavevectors. If the refractive index changes only along the depth z of the sample, the reflectivity is obtained by solving a one-dimensional wave equation, which for non magnetic materials takes the simple form:

$$\varphi''(z) + [Q^2/4 - 4\pi N(z)]\varphi(z) = 0 \qquad (2)$$

Substituting N(z) with a histogram of layers with constant potential, it can be shown[6] that the wavefunction is:

$$\varphi = A \exp(ik_z z) + B \exp(-ik_z z) \qquad (3)$$

in each layer of constant momentum k_z, with coefficients A and B obtained by imposing continuity of matter and flux at each layer boundary. The value of $|B|^2$ calculated at z=0

(the surface boundary) is the reflectivity. Similar expressions are obtained for polarized neutrons reflected by uniaxial ferromagnets, substituting N(z) with $\{N(z)\pm M(z)\}$.

Fig.3 shows the spin-dependent reflectivity calculated for a ferromagnetic film of iron on a (non-magnetic) chromium substrate. The oscillatory pattern is due to the interference of waves reflected by top and bottom of the iron layer, whose thickness is determined quite accurately from the oscillation period. In Fig.3 the iron magnetization is assumed uniform across the layer. If that was not the case, because of a weaker magnetization at the iron surface, the (+) and (-) reflectivities would become identical for increasing k_0.

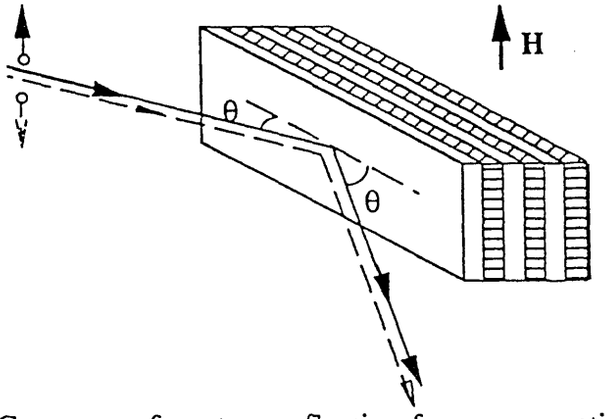

Fig.2 Geometry of neutron reflection from a magnetic multilayer. Full and dash rays refer to neutrons polarized parallel and antiparallel to an external magnetic field H.

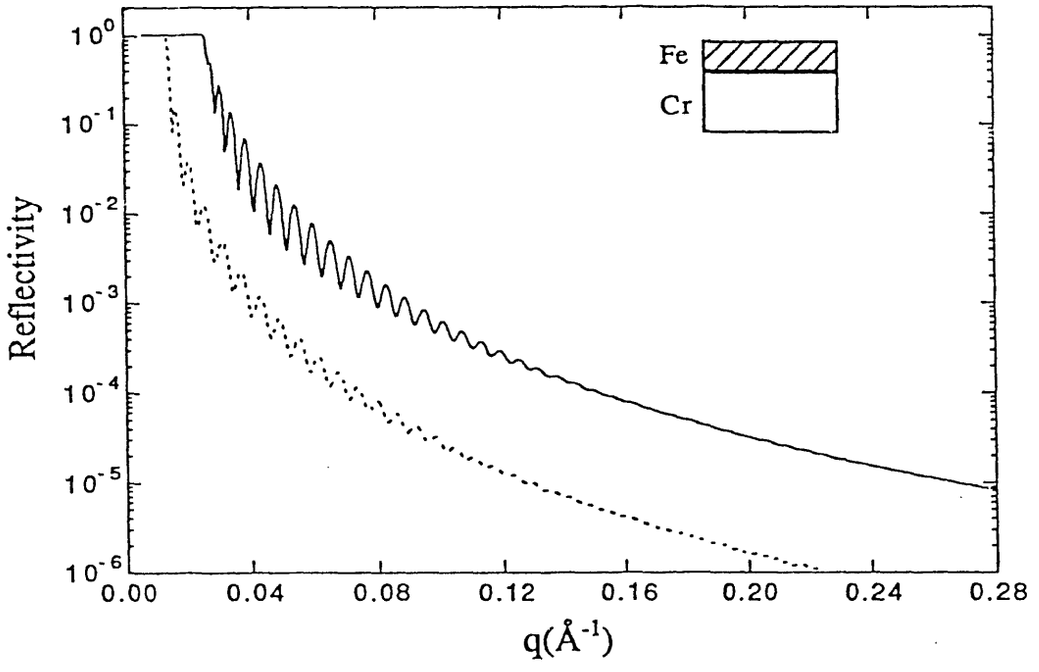

Fig.3 Spin-dependent reflectivity calculated for a magnetically saturated layer of iron, 100 nm thick, on chronmium. The magnetization of iron is assumed to be 1700 gauss, as in the body centered cubic solid. Full line: neutron spins parallel (+), dotted line: spin antiparallel (-) to the magnetization.

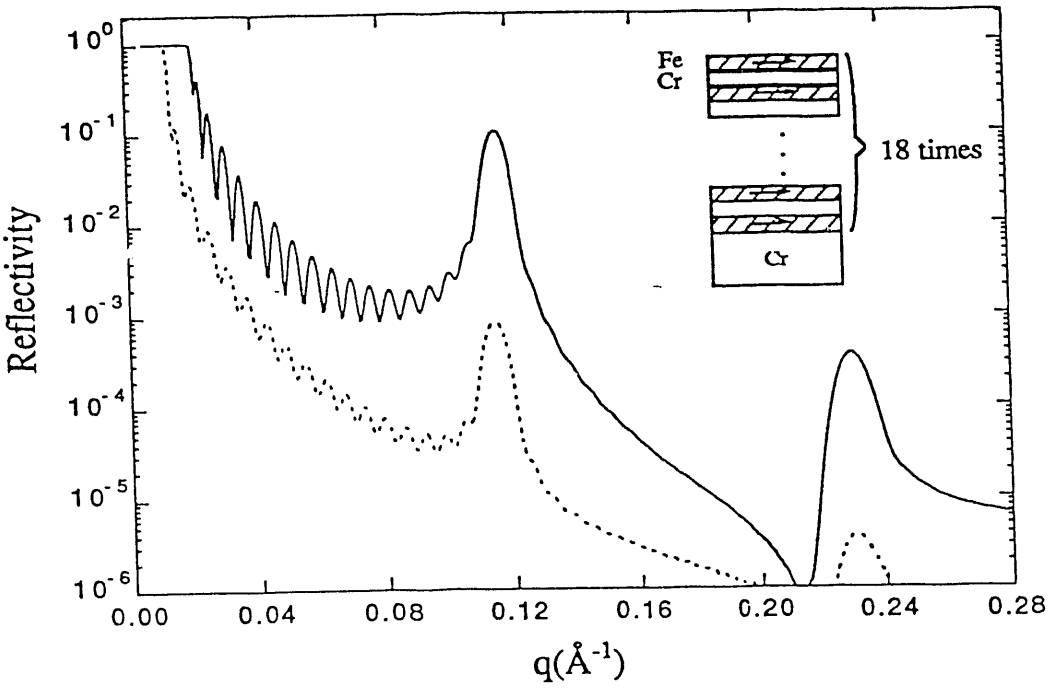

Fig.4 Calculated reflectivity of a multilayer, composed of 18 sanwiches of Fe (3.0 nm) / Cr (2.5nm) for a total thickness of 99 nm. The single layers are ferromagnetic, as in solid b.c.c. iron, and are magnetized parallel to each other.

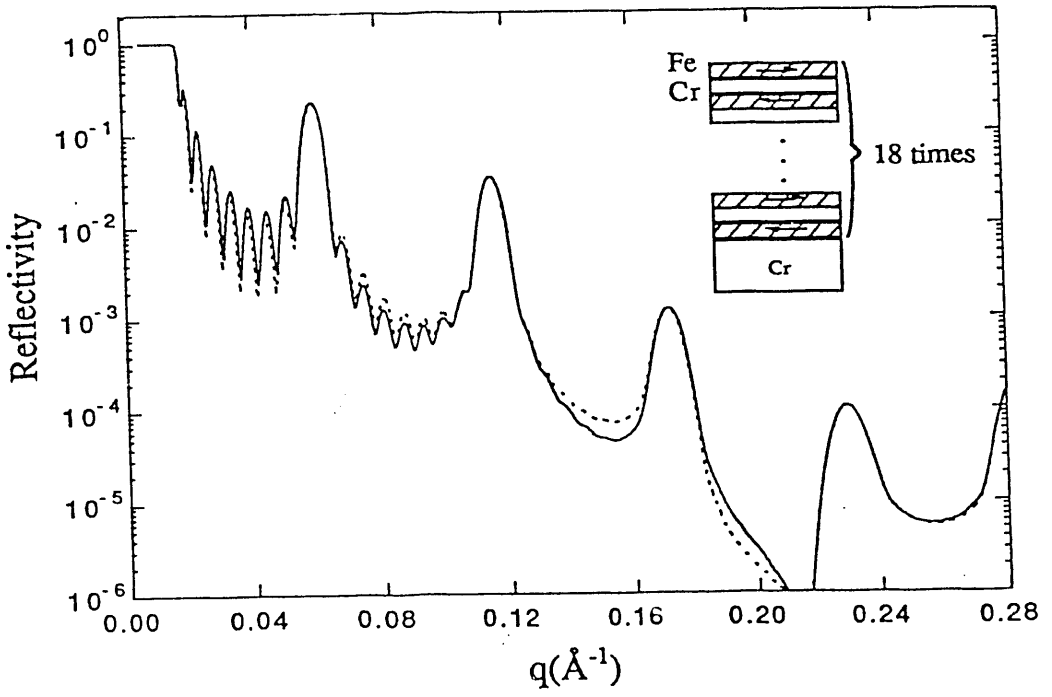

Fig.5 Calculated reflectivity of the same multilayer of Fig.4, except that here successive iron layers are magnetized opposite to each other.

In Fig.4 is presented the reflectivity due to a stack of n ferromagnetic iron layers, of thickness d_f, interleaved with chromium spacers (thickness d_s). As in Fig.3, the high frequency oscillations are due to interference of waves from the surface and the chromium substrate, at a depth $D=n(d_f+d_s)$ from the surface. Visible are also the two "Bragg" diffraction peaks, corresponding to a period d_f+d_s. The Bragg peaks are strongly spin-dependent because all the layers are magnetized in the same direction: the magnetic and crystal periodicities are identical. We will call synthetically this configuration as F. If instead successive layers have opposite magnetization (AF configuration), the magnetic period is twice the crystalline period. As a consequence the reflectivity shows additional Bragg reflections of magnetic origin, and basically is spin-independent (Fig.5).

In summary, polarized neutron reflection can determine directly the magnetic periodicity and compare it with the chemical periodicity. However, the measurement contains a wealth of information that reaches far beyond this point[7]. As observed in Fig.5, the reflectivity remains slightly spin-dependent even for a system with zero net magnetization. This comes about because the neutron probes the multilayer starting from the surface, and thus its response is biased by the relative orientation of the magnetization of the surface layer. To have a spin independent reflectivity the sample must contain at least two kind of antiphase domains with equal population. Polarized neutron reflection can also determine the *direction* of the magnetization at any depth in the sample, in reference to an external magnetic field. This is because the external field provides the quantization axis for the neutron spins: if the neutrons encounter along their path magnetic fields pointing in different directions, their spin precess around the local magnetization axis, and the exiting neutrons have polarization different from that at the entrance. With this considerations in mind, the experimental reflectivity can be fitted only when the correct model for the chemical and magnetic depth profile is used[8]. If the magnetic and chemical layers are not uniform across the surface, part of the neutrons are scattered in directions other than that of specular reflection.

3. Ferromagnetic thin films

Ferromagnetic films, ranging from a thickness of a single atomic plane to a fraction of a micron, are obtained by sputtering or vapor deposition of iron, cobalt, nickel or gadolinium on different substrates. Only for film thicknesses below a few nanometers the film magnetization is significantly altered from the bulk value, in size, direction of magnetization and even type of magnetic order. These new properties are the result of a complex set of circumstances. To start with, the magnetization of the surface layer is different from the bulk value even for conventional magnetic metals. On the other hand, free standing films, one atomic plane thick, exhibit magnetic moments larger than the bulk and tending toward the free atom values. At least this is the result of calculations; experimentally, films have to be deposited (at least, on earth) on a substrate, which perturbs the magnetization of the proximate layer on two accounts. The overlayer tends to mimic the crystalline structure of the substrate, even when the mismatch between the two cells is significant. The expansion (or compression) might create havoc with the coupling of the magnetic electrons: two spins occupying the same space are paired with opposite spins according to Pauli exclusion principle. When both magnetic film and substrate are metals a transfer of electrons can also take place. Numerous *ab initio* calculations have been made also for epitaxial films; Table 1 shows the magnetic moments predicted for one-atomic-layer-thick metals on several substrates[9,10].

While it is relatively easy for theorists to model ideal monolayers in computer

simulations, it is a tremendous challenge for experimentalists to grow clean systems in the laboratory. For instance, Cu, Ag and Au single crystals should make good substrates, on the ground that their *d* band is filled and not polarizable by the magnetic moments of the overlayers. However the same substrates have a tendency to form a composite buffer layer resulting either by intermixing or segregation. The magnetic properties of the resulting system depend dramatically on the mode of deposition or the substrate temperature. A good example of how complex the resulting phenomenology could be is provided by a set of experiments[11] addressed to measure the magnetic anisotropy of face-centered-cubic iron on copper. The direction of the magnetization is in the plane of the film, or tilt, or perpendicular to it according to the growth temperature and the film thickness. Difficulties of different kind arise when metallic substrates are made of open *d* shells. There, band hybridization is most likely to occur: perhaps the most spectacular result of this effect is the ferromagnetism induced in a palladium substrate covered with iron[12].

Table I. Calculated magnetic moments for monolayer of 3*d* metals, in Bohr magnetons (after ref.9). These are compared with the experimental values for the solid metals.

Metal	Magnetic Moment in Solid(μ_B)	Type of Monolayer	Magnetic Moment in monolayer(μ_B)
V	<0.1	V/Au(001)	1.75
		V/Ag(001)	1.98
Cr	0.45(AF)	Cr/Vacuum	4.12
		Cr/Au(001)	3.70
Fe	2.2	Fe/Vacuum	3.20
		Fe/Ag(001)	2.96
		$Fe/W(110)_u$	2.97
		$Fe/W(110)_r$	2.18
Co	1.78	Co/Cu(001)	1.79
Ni	0.6	Ni/Ag(001)	0.57

u: unrelaxed lattice; r: relaxed lattice

A fair amount of work has been carried out on semiconducting or insulating substrates. These have the obvious advantage of not exchanging electrons with the metallic epilayers. Also, in some case they can be grown in highly controlled conditions and be atomically flat. Thin films of iron[13] and cobalt[14] have been deposited on GaAs, and large variations of both the size and the orientation of the ferromagnetic moments have been found for thicknesses less than 5 nm. These have been attributed in large part to the mismatch in lattice spacing of the substrate and the epilayer. Another interesting substrate is MgO because, in the (100) orientation, its lattice dimensions closely match those of body-centered-cubic iron. Fig. 6 shows the charge and the momentum density calculated[9] for iron deposited onto a (100) substrate of magnesium oxide. The predicted magnetic moment for iron, 3.07 μ_B, approaches the value for the free standing Fe monolayer. Its difference from the bulk value should become quite apparent in a quantitative measurement.

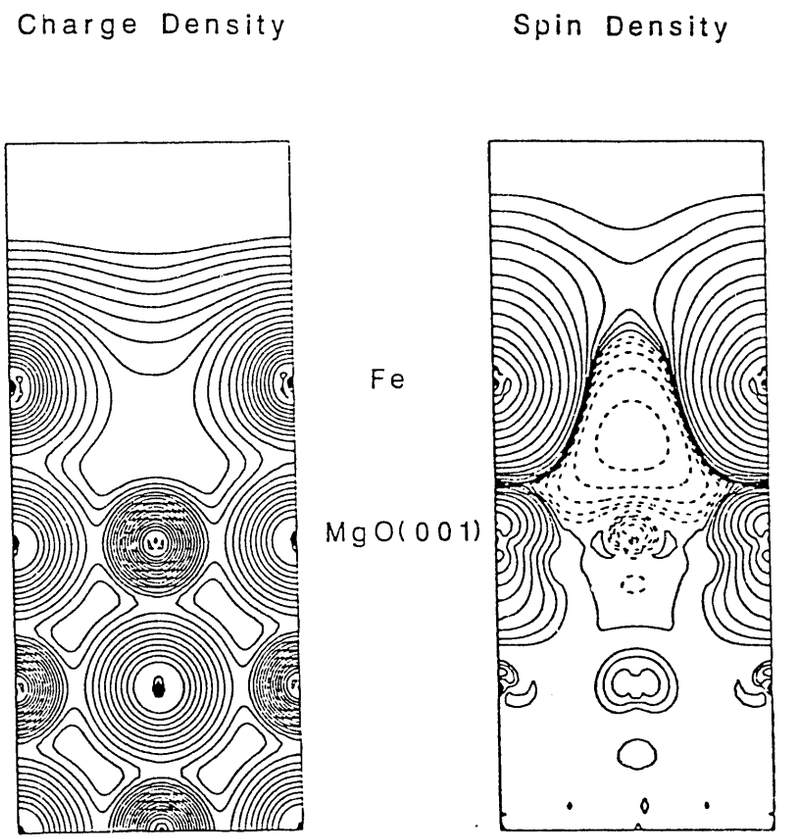

Fig.6 Charge and spin density calculated for a single atomic plane of iron on MgO (from ref. 9)

To check this prediction, polarized neutron reflection measurements were taken[15] for a number of films with progressively thinner Fe. In Fig. 7 are presented the results for a film of iron, four atomic planes thick, on MgO. The iron has been covered with 10 nm of gold for protection in these *ex-situ* measurement. The remarkable spin-dependence of the reflectivity shows that the magnetization remains sizeable even at these iron thicknesses.

The gold coating actually enhances the magnetic signal as a quarter-wave plate[16]. The polarization has the form:

$$R^+/R^- = 1 + 8k_0 \sin 2d_{Au}k_{Au} \times (M_{Fe}d_{Fe}/N_{Au}) + \text{higher order terms} \qquad (4)$$

where k_{Au} is the neutron's momentum in gold and d_{Fe} is the iron thickness. By fitting Eq.(4) to the data, and from the independent knowledge of the film thickness, the Fe magnetization is obtained quantitatively. At low temperature all samples were found to be ferromagnetic. Fig. 8 shows the ordered magnetic moment per iron as obtained from polarized neutron reflection data for a two-monolayer thick sample. In plane saturation is achieved only in a magnetic field of a few kiloOersted; the magnetic moment per iron is perhaps larger than in the solid but lower than predicted for a thin film.

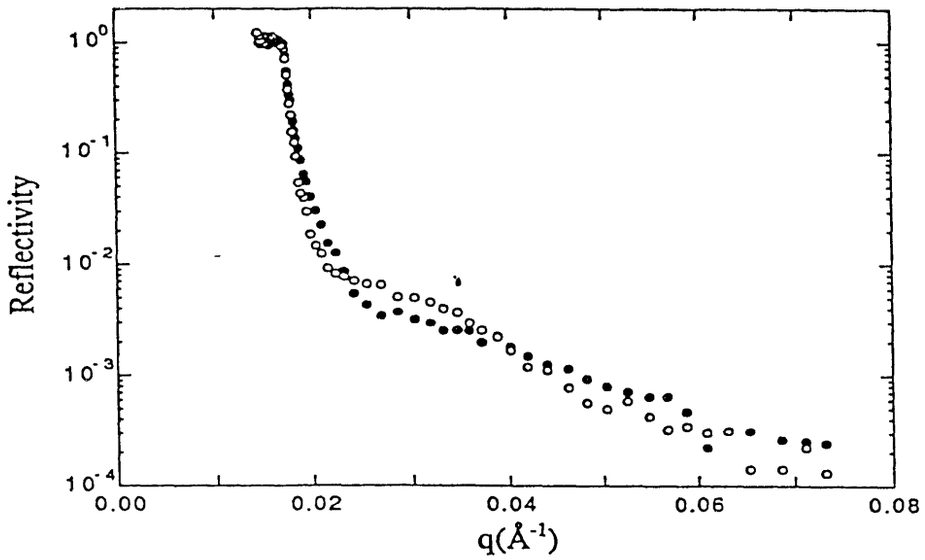

Fig.7 Spin-dependent reflectivity for a sample of iron, 4 monolayer thick, on MgO

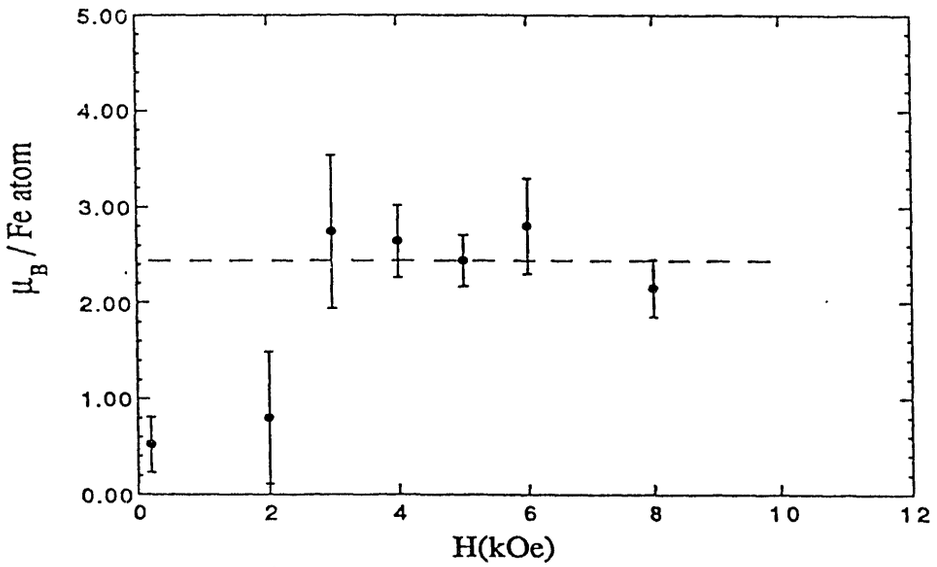

Fig.8 Low temperature magnetization of a iron film (2atomic planes thick) on MgO

4. Biasing the magnetism of a thin layer

Thin films can be magnetically biased when inserted with a certain procedure in an antiferromagnetic medium. For instance, the magnetic hystheresis loop of the permalloy film of Fig. 9 is not centered around the origin but rather around H_b - the bias field[17]. At first sight it might seem paradoxical that effects of this kind occur for a ferromagnet, since the magnetic energy is proportional to B^2 and invariant under field reversal.

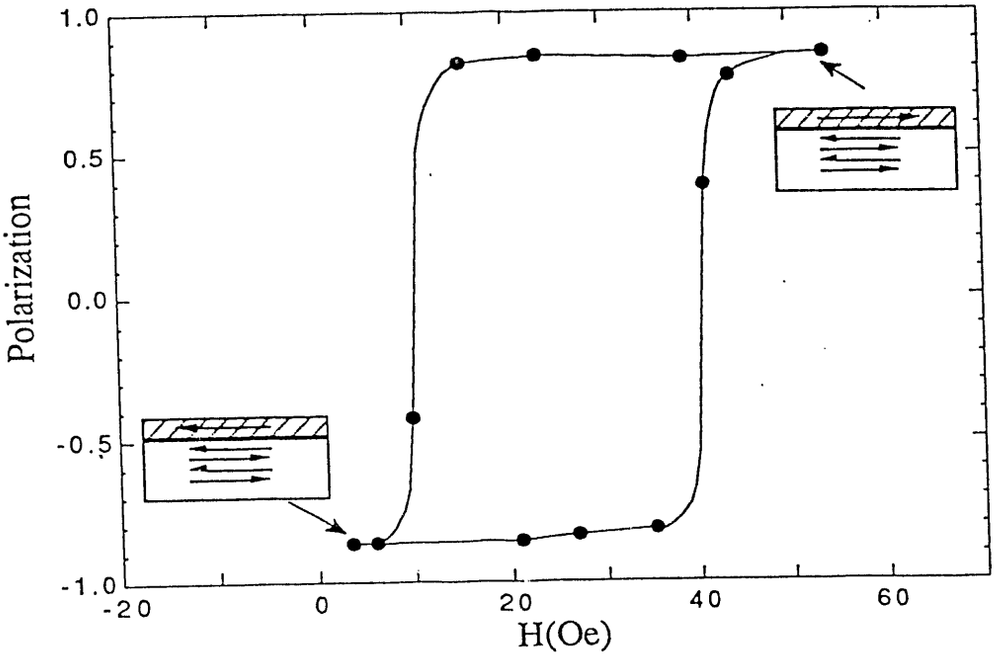

Fig.9 The magnetization of permalloy on $Ni_{0.5}Co_{0.5}O$. The magnetic signal is obtained from polarized neutron reflection measurements. At the right and left: cartoons of the proposed magnetic structure.

The phenomenon of unidirectional anisotropy has been discovered in the late 1950's in coupled ferromagnetic-antiferromagnetic systems such as Co/CoO and later also in spin glasses. With the development of the modern techniques of thin film preparation, controlled and reproducible bias has been obtained in couples permalloy/FeMn, permalloy/TbFe etc. There are several ways to introduce the permanent magnetic bias during the film deposition. For instance, in the case of the couple comprising the ferromagnetic alloy $Fe_{0.81}Ni_{0.19}$ (permalloy) and the antiferromagnetic $Ni_{0.5}Co_{0.5}O$, the film is evaporated in presence of a magnetic field, and the film retains memory of its direction. The general requirement is that the antiferromagnet orders at lower temperature than the ferromagnet[18].

Since its discovery, the unidirectional bias has been attributed to the preferential coupling of the moments of last atomic plane in the ferromagnet with those of the first atomic plane in the antiferromagnet. Suppose that this coupling is ferromagnetic (insert at the left, Fig.9). If the magnetic field H is reversed, the ferromagnet's magnetization is reversed, but the two layers at the interface have now opposite magnetization (insert at the right, Fig.9). H_b is the exchange bias field necessary to stabilize the unfavorable coupling across the interface .

This explanation is not entirely satisfactory. Simple estimates based on reasonable values of the exchange constants yield a value for the exchange anisotropy which is about 100 times larger than that observed experimentally. Various mechanisms have been proposed to account for such unexpectedly low value of the exchange anisotropy. Unfortunately, these are complex systems - for instance, the magnetic structure of $Ni_{0.5}Co_{0.5}O$ remains somewhat controversial. However, the fact that the magnitude of the exchange anisotropy found for film couples prepared by different researchers under different preparation conditions is similar strongly suggests an intrinsic mechanism for the

reduced magnitude. Possible mechanisms include domain-wall formation in the antiferromagnetic layer or the presence of massive amounts of imperfections at the interface[19]. For example, terraces at the interface or intrusions of permalloy in the oxide layer may cause the averaged exchange anisotropy to be reduced. To explain why the effect is not averaged completely to zero it was proposed[20] that the presence of random interface roughness gives rise to a random field acting on the interface spins. The antiferromagnet then breaks up into domains of size determined by the competition of exchange and in-plane anisotropy, and this size sets the scale for averaging the random field.

The applicability of these models can be checked in principle by polarized neutron reflection: at the saturating fields $+H_s$, $-H_s$ the net magnetization at these two states is exactly the same, however the sequence of layer magnetism experienced by neutrons in their flight path is different, and should cause some spin dependence in the reflectivity at $k_0>0$. Careful spin-dependent reflectivities taken at $+H_s$, $-H_s$ at first look appear identical. Minute differences (at present barely above statistics) suggest a slightly uncompensated modulation in the antiferromagnetic layer for a thickness of 30 Ångstroms. In summary, the phenomenon of magnetic bias is not yet fully understood; however empirically it is possible to produce thin films having the desired bias.

5. Magnetic coupling in multilayers

The magnetic properties of metallic multilayers, formed by interleaving ferromagnetic layers with metallic spacers of a given thickness, have been intensively studied in the past decade, following the development of reliable and controlled deposition techniques. Is it possible to describe in a unified way the magnetic interactions between different layers? Four decades ago[21,22] Rudermann, Kittel, Kasuya and Yosida made a basic proposal, concerning the interaction of two spins (nuclear or electronic) connected by an electron bath. The proposed mechanism, since then called RKKY, has been recently adapted to the systems at hand. The exchange interaction is expressed by:

$$F(x) = (1/x^4)(x\cos x - \sin x) \tag{5}$$

with

$$x=2k_FR \tag{6}$$

In eq. (5), k_F is the Fermi momentum and R the distance between the two spins. Eq. (4) shows that the coupling between the two spins oscillates as a function of their distance from positive to negative values and then to positive again, with interaction maxima which are progressively damped.

The first materials to exhibit oscillatory magnetic interaction were epitaxially grown gadolinium/yttrium superlattices[23]. Gd, with its half shell of *4f* electrons, is a strong ferromagnet even when thinned down to a few atomic planes. In superlattices with yttrium, subsequent Gd layers were found to have parallel magnetic moments (F) when the yttrium spacer is six atomic planes thick; antiparallel moments (AF) for a Y thickness of ten atomic planes. To apply the RKKY scheme Yafet[24] simplified the system, assuming single plane arrays of gadolinium embedded as impurities in a Y matrix. In spite of the seemingly crude approximation, the exchange coupling curve calculated as a function of the Y thickness shows indeed a F coupling at 6 Y atomic planes, and an AF coupling at 10 atomic

planes.

The seminal work on superlattices of Gd/Y opened the possibility of engineering entirely novel magnetic materials. Is this possibility limited to exotic metals, or to very sophisticated techniques of deposition? This is not the case. In a series of recent experiment, it was shown that films of iron/chromium, both produced by molecular beam epitaxy *or* sputtering, have a saturating field strongly dependent on the chromium thickness[25,26,27]. The saturation field oscillates (as shown in Fig.10), and it was inferred that the maxima occurred for a compensated ground state, where successive iron layers have opposite magnetization. The only magnetic structures are of the F and the AF kind, and the latter configuration is destroyed when sufficient magnetic field is applied. This description has been confirmed by reflectivity measurements with polarized neutrons[28,29] (Fig.11).

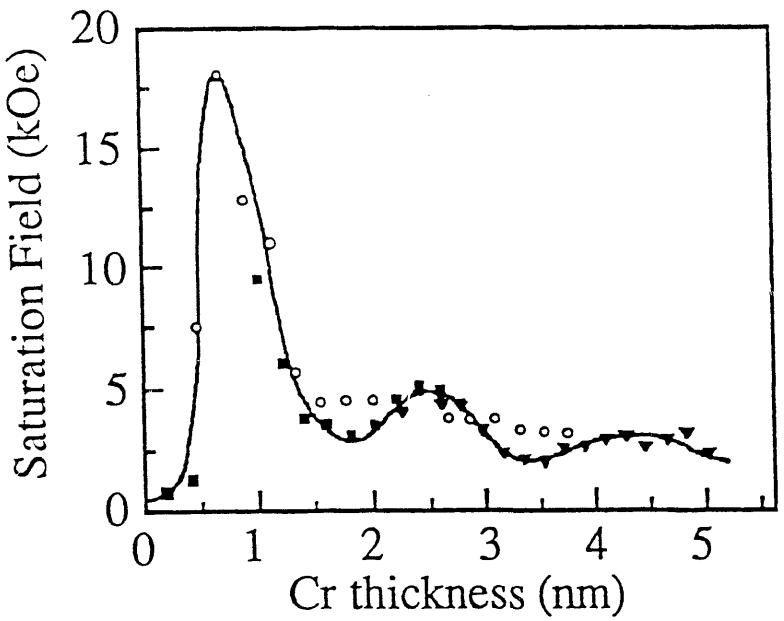

Fig. 10 Saturation magnetic field for Fe/Cr multilayers. The thickness if iron is kept to 20 Å while that of Cr is varied. Full dots: iron deposited at 40 C; open dots: deposited at 125 C (from ref. 27)

The oscillatory nature of the magnetic coupling in multilayer ferromagnetic seems now much more common than previously thought. Once chosen the spacer, the coupling between layers of iron, cobalt, nickel is quite similar. Table II shows the properties of cobalt (perhaps the most studied of the ferromagnetic elements) in multilayers with other *3d, 4d, 5d* materials[30]. Perhaps its most remarkable feature is that the period of oscillations P is (with the exception of chromium) approximately constant across the table.

Can the oscillation of magnetic coupling in these multilayers be explained by a RKKY-type interaction? In its most simple form, the period of the RKKY oscillations is π/k_F, or several times shorter than experimentally observed. To account for the discrepancies, several theoretical approaches have been put forward, in which the RKKY oscillations were deeply modified by the introduction of surface roughness. If roughness is present, the effective interaction must be obtained by averaging over magnetic configurations with different spacer thickness. The first paper adopting this view[31]

considered in particular the Fe/Cr system. Starting from basically the same picture successfully proposed by Yafet[24] for the rare earths, it points out that the coupling of the iron moments with the conduction electrons of Cr comes from two sources: the Coulomb exchange and mixing (or hybridization). The mixing interaction, zero for the free electron

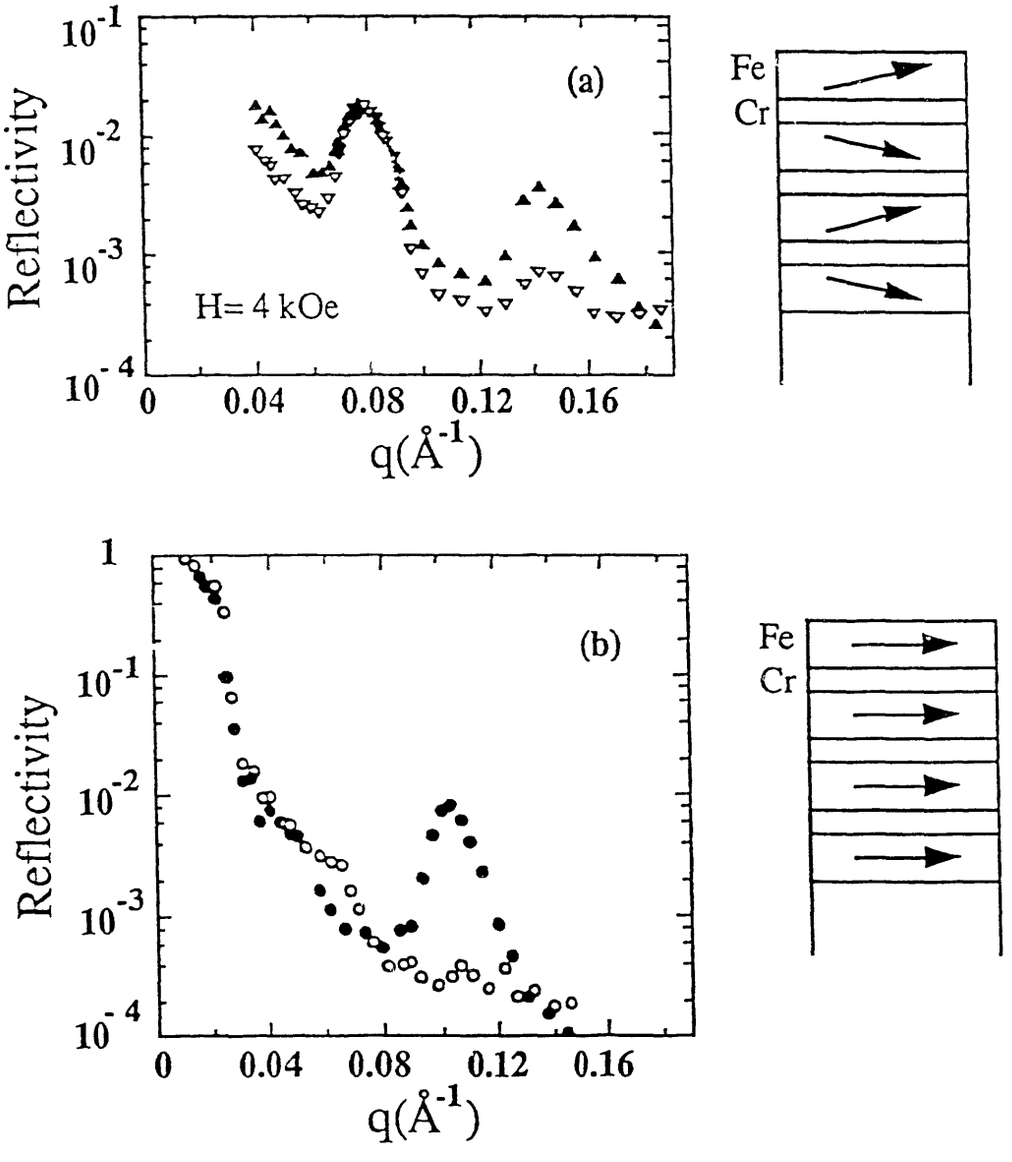

Fig. 11 (a) Spin-dependent neutron reflectivity of $\{Fe(32Å)/Cr(10Å)\}_{20}$ in a magnetic field of 4 kOe. Solid triangles: spin +. Open triangles: spin -. The magnetic moments of the Fe layers are canted, and the AF component gives rise to the spin-independent peak at q = 0.08 Å^{-1}, the F component to the peak at q = 0.143 Å^{-1}. (b) Effect of the magnetic field on the AF peak of $\{Fe(20Å)/Cr(10Å)\}_{20}$. Solid dots: spin-averaged reflectivity at H=4kOe. A field of 14 kOe saturates the sample, causing the disappearance of the AF peak at q=0.11 Å^{-1} (open dots)

Table II. Magnetic properties of Co/TM multilayers. For each Transition Metal is indicated: A_1, the spacer thickness (in Ångstroms) for which the AF coupling is strongest; J_1, the energy of the coupling (in erg/cm^2); ΔA_1, the thickness range for AF coupling (in Ångstroms) and the period P of the saturation field oscillations. In the cases "*", only one AF coupled spacer-layer thickness was observed (after ref. 30).

V		Cr		Mn		Fe		Co		Ni		Cu	
9	3	7	7									8	3
0.1	9	.24	18									0.3	10
Nb		**Mo**		**Tc**		**Ru**		**Rh**		**Pd**		**Ag**	
9.5	2.5	5.2	3			3	3	7.9	3				
.02	*	.12	11			5	11	1.6	9				
Ta		**W**		**Re**		**Os**		**Ir**		**Pt**		**Au**	
7	2	5.5	3	4.2	3.5			4	3				
.01	*	.3	*	.41	10			1.85	9				

Element	
A_1	ΔA_1
J_1	P

case and small for the rare-earth-yttrium layered structures, is sizeable for *3d* metals. The apparent period in Fe/Cr multilayers stems from the competition between Coulomb exchange and the hybridization of the chromium electrons close to the Fermi surface. The RKKY oscillations present in the Coulomb exchange term are smeared out by roughness. The calculated period fits well that found experimentally for Fe/Cr. An alternative approach[32] consists in an extension of the general theory of RKKY exchange. Calculations for Cu, Ag and Au spacers showed that large periods and multiperiodic oscillations are natural consequences of the discreteness of the spacer thickness and of the moment distribution within the ferromagnetic layers , even within the free-electron approximation. The periodicity of the oscillations for noble metals was found to be significantly different from π/k_f and dependent on the epitaxiality. Even in this case, however, fitting with the experimental periods was obtained only after introducing a significant roughness at the interface. In general, the effect of the roughness is that of suppressing the short period oscillations: acting as a low-pass filter, roughness leaves only the weak, long period oscillatory coupling.

The importance of roughness in determining the periodicity of magnetic coupling in multilayers has been directly confirmed in a beautiful experiment[33]. An Fe(100) single crystal whisker was covered with a wedge-shaped layer of evaporated chromium (Fig.12), and this in turn was covered by a monolayer of Fe. The experiment consisted in measuring the direction of magnetization of the top Fe layer as a continuous function of the Cr spacer thickness. The reference magnetization is that of the substrate Fe wiskers, where two domains are present, corresponding to the lower and upper bands of the two panels

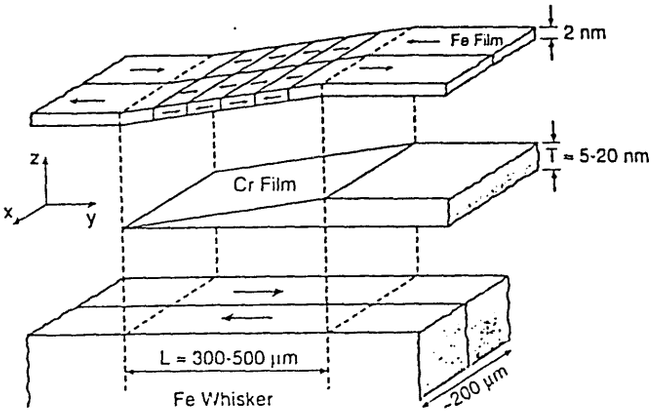

Fig. 12 Exploded view of the single crystal iron substrate, the evaporated Cr wedge, and the Fe overlayer. The arrows in the Fe show the direction of the magnetization in each domain. The actual wedge angle is of the order of 10^{-3} degrees (from ref.33)

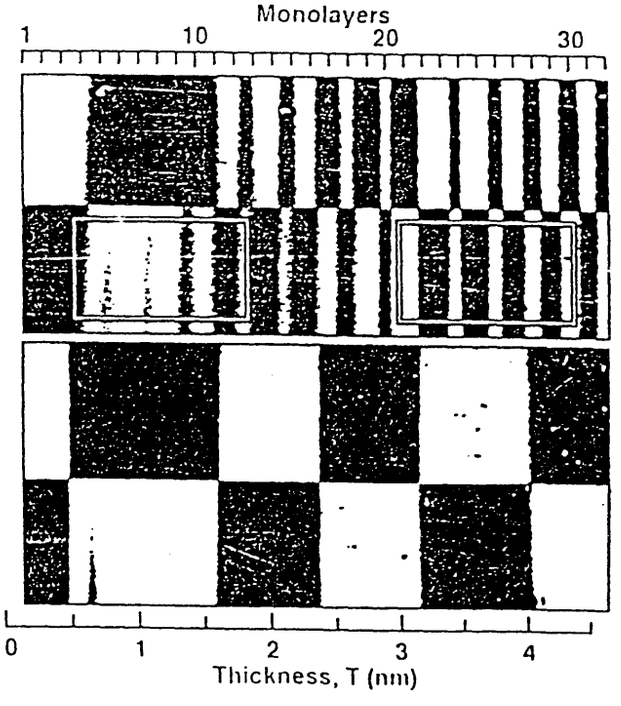

Fig.13 SEMPA images showing the magnetization of the upper layer of Fe in Fe/Cr/Fe wedge sandwiches. Upper panel: sandwich grown on a hot substrate; lower panel: substrate at room temperature. The two horizontal stripes in each panel correspond to opposite magnetization state of the Fe substrate. Each panel represents an area of approximately 0.3 x 0.3 mm (from ref.33).

displayed in Fig.13. The magnetization of the top Fe layer was measured by scanning electron microscopy with polarization analysis (SEMPA): the black and white spots of Fig.13 correspond to opposite polarization. Also, the crystal perfection was monitored by reflection high-energy electron diffraction (RHEED). Two samples were grown on substrates at different temperatures. For the sample grown at room temperature the oscillation of the surface Fe magnetization had the same period as in sputtered samples. At the same time, the RHEED pattern of chromium showed the presence of some imperfections. For the sample grown on a hot substrate (250 C) the RHEED pattern indicated an almost ideal epitaxial growth, while SEMPA showed a magnetization oscillating with the period expected for RKKY interactions (Fig.13).

Admittedly the approach to produce multilayers of controlled magnetic properties is still semiempirical. However, the ongoing research is stimulated also by other aspects of their phenomenology that are decisively intriguing. The magnetoresistance, or the difference in electric resistance in zero magnetic field and in a saturating field, is very large or "giant" for those multilayers in an AF ground state[26,27,28]. While in conventional systems magnetoresistive effects amount to only a few percents[34], the resistance of some multilayers changes by as much as 80%. It becomes feasible to read the magnetic state with a simple electric measurement. Fig. 14 shows the behavior of the magnetoresistance for a sputtered film of Co/Cu as a function of the applied field[35]. The maximum of the magnetoresistance, as a function of the spacer thickness, follows the same oscillatory behavior as the saturating magnetic field[27]. The presence of an AF state seems necessary for a large magnetoresitance, but is not sufficient: epitaxially grown multilayers exhibit usually much less magnetoresistance than samples grown by sputtering[35]. Early speculations[26] linked the effect to the presence of point defects at the domain walls which would distort the local spin configuration and thus scatter electrons. Recent measurements[36] revealed the presence of an unusually large amount of relatively small magnetic domains in the AF region, or of an unusually large amount of magnetic walls which might be responsible for the scattering of the electrons. This evidence is contained in the neutron scattering pattern shown in Fig. 15.

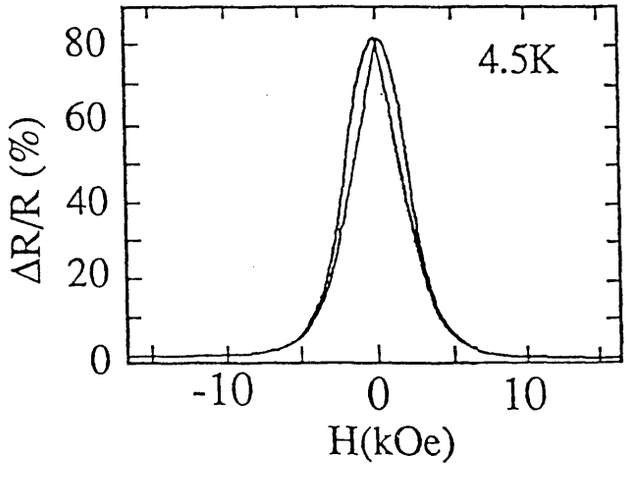

Fig.14 Magnetoresistance of a sputtered Co/Cu multilayer at low temperature, referenced to the saturated state. A slight hysteresis is observable. ΔR/R decreases considerably by warming the material (from ref.35)

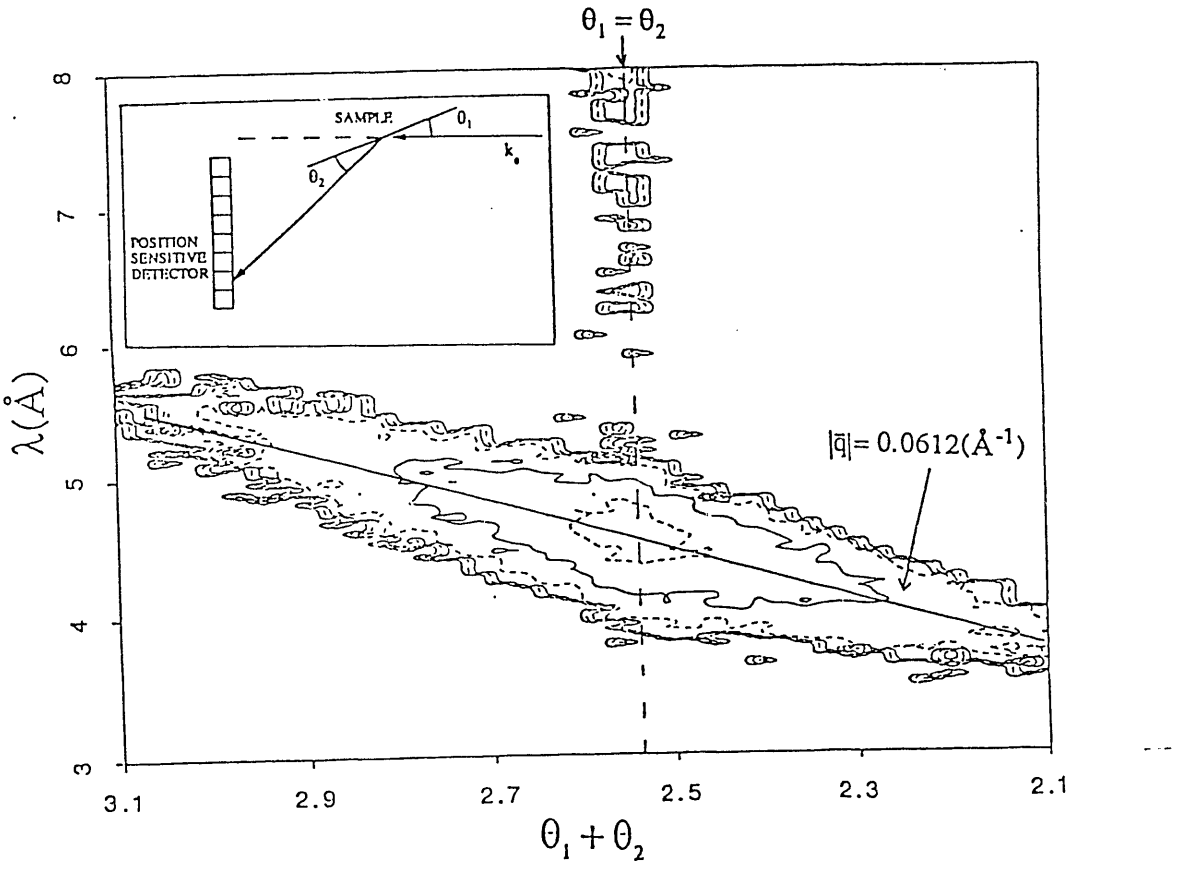

Fig.15 Intensity contours for the neutron reflectivity and diffuse scattering from antiferromagnetic domains of a Co/Ru sample. The insert shows the scattering geometry. Neutrons are reflected only on the line $\theta_1=\theta_2$; the large scattering at $q= 0.0621$ Å^{-1} (the position of the AF peak) indicates that the AF domains width is only a small fraction of a micron .

6. Surface-induced phase transitions

Magnetic multilayers form a novel kind of magnetic system, made of strongly ferromagnetic layers with weak (and tunable) coupling. Their phase diagram is quite distinct from those found for magnetically ordered solids: even a weak magnetic field changes the magnetic configuration from AF to aligned. As it is well documented for solids[37], a phase transformation may be initiated at a surface and then propagate in the bulk. Similar effects might take place in multilayers. Actually a surface driven phase transformation has been recently proposed for a model multilayer system[38], with interlayer interaction somewhat different from those discussed up to now. While this system was created as a mean field model, experimental verification of its predicted phenomenology starts to be accumulated[39,40].

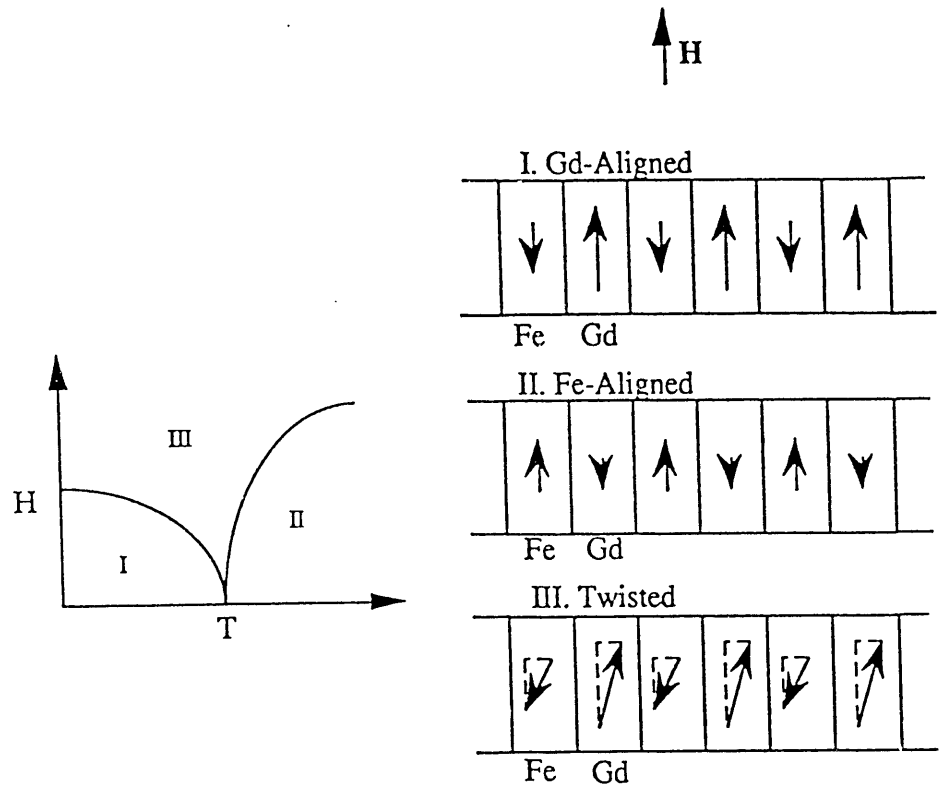

Fig.16 Phase diagram for the "solid" Gd/Fe multilayer. Not shown are the disordered phase (high temperature) and the field-aligned phase (high magnetic field). After ref.38.

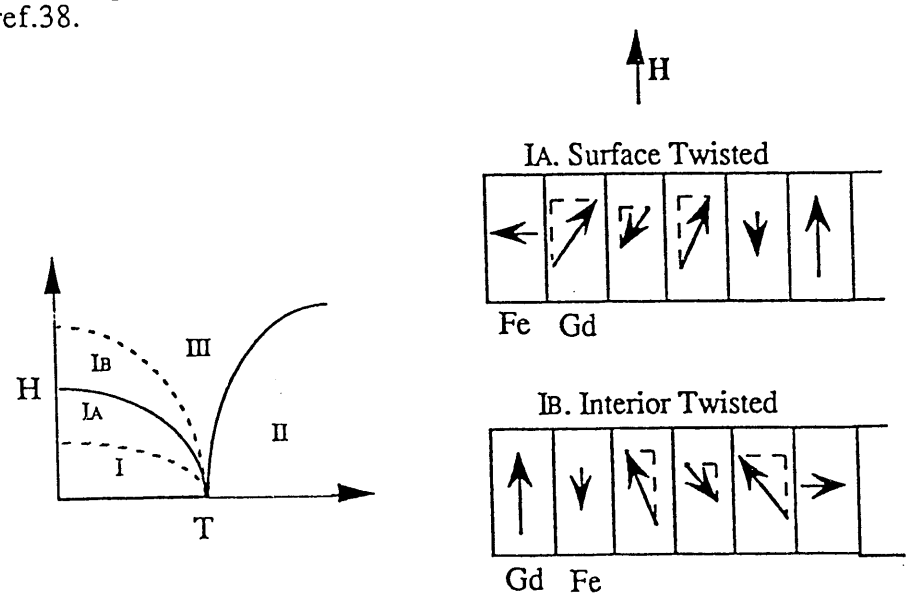

Fig.17 Effect of the surface on the phase diagram of Gd/Fe. If the surface layer is iron, a twisted state is induced at the surface in field much lower than the bulk. The penetration depth can be fairly large. In contrast, Gd at the surface hinders the transition (after ref.42).

Suppose that a multilayer is composed of couples of two ferromagnetic components with opposite alignment. For simplicity these component will be named "iron" and "gadolinium" because layers of these two elements have indeed similar properties. Also, the magnetic moment at saturation per iron ($2.2\mu_B$) is quite different from that of Gd ($7\mu_B$). At zero temperature the couple magnetization can be -depending on the relative thickness of the two layers- exactly compensated, or Fe aligned, or Gd aligned. In the last case an interesting evolution of the magnetization takes place when the temperature is raised. The magnetic interaction between Gd atoms $J_{Gd\text{-}Gd}= \epsilon$, is in fact much weaker than the ferromagnetic interaction in iron $J_{Fe\text{-}Fe}=1$ or the antiferromagnetic one between the two materials $J_{Gd\text{-}Fe}=-1$. Therefore, by raising the temperature, the effective ordered moment of Gd is thermally lowered more than that of Fe. The system becomes compensated, and then Fe aligned. The phase diagram is shown in Fig.16.

The onset of an external magnetic field creates additional phases. For small magnetic field the Zeeman energy term is sufficiently weak and minimization of the exchange energy generally favors the aligned state of zero field. The magnetization is constant in this region. Above a critical field this arrangement is broken, to form a new structure which has been called[38] "twisted". Also this structure is sketched in Fig.16. It can be observed that the Gd and Fe components perpendicular to the field are exactly compensating each other, and, if switched, they give rise to a second magnetic structure, which is exactly equivalent to the former. The representation of the magnetic moments given in Fig.16 is quite simplified, lumping together all the spins in a single layer. In the detailed model proposed[38] those spins are spread in a narrow angle fan. Further, at finite temperature the size of Gd (and Fe) moments is also modulated. In the twisted state region the magnetization grows more or less linearly with the field[41].

The field-induced transition from the Gd aligned to the twisted state is deeply influenced by the presence of the surface. By surface here are meant the outmost layers of the stacking, respectively at the surface and facing the substrate. The transition depends crucially on the nature of the outmost layer, as can be seen readily with a plausibility argument[42]. In the Gd-aligned phase the Fe spins are antiparallel to the external field. Fe spins in the interior are strongly held antiparallel to the external field by the antiferrromagnetic coupling to the Gd spins on both sides of Fe. In contrast, Fe spins at the surface are not as strongly fixed since they have Gd on one side only, and tend to turn in a magnetic field lower than the critical field for the bulk transition. Fig.17 shows the open fan arrangement induced by the terminating the multilayer with Fe. If the multilayer were terminated with Gd, the opposite effect would take place: the twisting would start in the interior, and its full display would be actually hindered by the presence of the Gd surface. The qualitative arguments reported here have been backed up by rigorous calculations, where the energy of the Gd surface spin wave mode has been calculated, and found to become soft at a finite applied field[42]. Its penetration in the bulk has been found rather large from the calculation.

This concludes our review of this new field, which might be called, with tongue in cheek, that of ersatz ferromagnetism. From the dates of the references, it can be seen that the field is new indeed; and aside from opening new venues, it might help viewing in a fresh light a classic but somewhat aged subject.

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