Oscillatory Interlayer Coupling and Magnetoresistance in Magnetic Metallic Multilayers

M. N. Baibich

Instituto de Física, Universidade Federal do Rio Grande do Sul, 91500, Porto Alegre, RS, Brasil

and

R. B. Muniz

Instituto de Física, Universidade Federal Fluminense, 24020, Niterói, RJ, Brasil

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The oscillations of the interlayer exchange coupling and the giant magnetoresistance effect in magnetic metallic multilayers have been intensively investigated both experimentally and theoretically. The current status of the theory and some specific experimental results concerning these systerns are briefly reviewed.

I. Introduction

Different metals can be deposited in consecutive layers to form what are generally called metallic multilayers. These systems rnay exhibit physical properties which are very different from those of their constituent materials. With the development of new experimental techniques and refined control in materials science, it is now possible to monitor the growth and characterize these structures within an atomic scale[1]. Careful control of growth can produce systems with new periodicities, effectively of low dimensionality, and rnay place elements in stable structural phases in which they are not usually found in nature. Such new boundary conditions rnay greatly alter the underlying electronic structure, and hence the properties of tliese materials[2]. The design of new materials requires a constant interplay between theory and experiment.

The magnetic properties of metallic multilayers composed by rnagnetic and non-magnetic metals rnay be used to produce devices of great interest for the magnetic industry. For example, the magnetic anisotropy at the interface between metals like Co/Pt and Ni/Pt[3] can favor perpendicular magnetization, which is useful for producing high-density storage medium. An applied magnetic field can cause big changes in the resistance of certain magnetic metallic multilayers, and this effect rnay be exploited to construct magnetic sensors. Early magneloresistive sensors were based on materials which shon approximately 2% change in resistance, but in some metallic rnultilayers the change can be as high as 100%. This unexpectedly high effect has been called giant rnagnetoresistance and was first observed in Fe/Cr mul:ilayers by Baibich et al[4].

It lias also been observed[5,6] that the coupling between the magnetic layers of metallic rnultilayers rnay be either ferromagnetic or antiferrornagnetic depending on the thicknesses of the spacer layers. The period, phase and magnitude of the oscillations of the interlayer coupling, as well as the magnetoresistance, depend on the rnultilayer constituent materials, and interface quality rnay also play a very important role.

Here, we shall concentrate on magnetic metallic multilayers composed by ferromagnetic transition metals separated by non-magnetic transition or noble metal ~ Our attention is devoted to two magnetic properties observed in these multilayers, namely the oscillations in the interlayer coupling and the giant magnetoresistance effect.

II. Samples and Experimental Methods

Careful control of multilayer growth requires a great deal of science and not less of art. Most of the evaporation methods traditionally used to prepare thin films rnay also be employed to produce multilayers. The essential problem in growing these structures is to have a good control of the thicknesses, interface roughness and chemical purity of the layers.

Initially, fairly sophisticated methods like the Molecular Beam Epitaxy (MBE) were used to prepare good quality samples. Later, it was found that some of the properties originally attributed to the single crystal structures obtained by MBE were also present in policrystalline samples. More recently, it has been shown that even the epitaxy of certain metallic multilayers can be obtained with standard electron beam physical deposition machines[7]. These much simpler, faster and economical methods of sample preparation increase the potential technological application of these systerns in the magnetic industry.

Multilayers with translational syrnmetry in the direction perpendicular to the layers are called superlattices. Policrystalline multilayered samples retain the

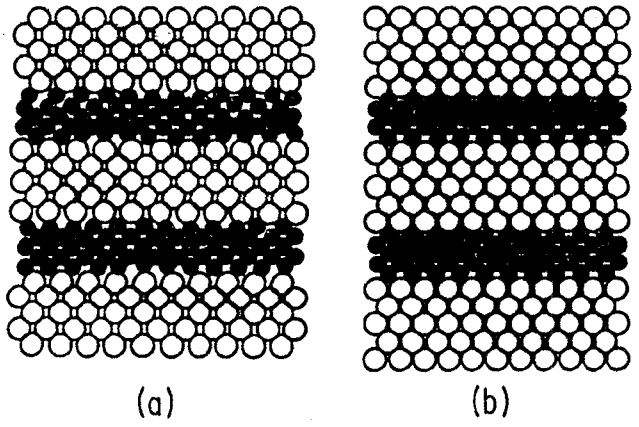

Figure 1.: Schematic representation of two-component superlattices (a) policrystalline sample (b) single crystal structure (after Draaisma ref. [8]).

compositional symmetry but not a unique crystal orientation, although they are usually predominantly textured in one direction. This is schematically shown in Fig. 1. One of the most useful and powerful methods for investigating the underlying symmetries of these systems is X-ray diffraction (XRD). Both small and large angles XRD provide information about the periodicity of the system and quality of its interfaces. Ideally, XRD patterns of a two component superlattice show either two intensity distributions (each centered around the corresponding Bragg peaks of the constituent materials), or a distinct intensity distribution around an average Bragg peak in the cases of large and thin layer thicknesses, respectively. These Bragg peaks are decorated by equally spaced satellite peaks associated with the chemical period of the bilayer. Deviations from the ideal situation can be analyzed with more refined models to obtain information about the sample texture, and roughness of the interfaces. Other experimental techniques, such as nuclear magnetic resonance (NMR) and Mössbauer spectroscopy look at changes in the hyperfine fields at suitable isotopes, and in some cases may be used to acquire additional information about the local structure and magnetic properties of the system.

III. Magnetic properties

The magnetic properties of layered structures can be studied by different experimental methods such as neutron scattering, Brillouin scattering, spin polarized low energy electron diffraction (SPLEED), ferromagnetic resonance (FMR), and by magneto-transport, magnetization and torque measurements. Ferromagnetic and antiferromagnetic alignments of the magnetic layers lead to different behaviors of the magnetization M as a function of an applied magnetic field H. The hysteresis loop shown in Fig. 2, where the remanent magnetization is hardly visible and the saturation field is

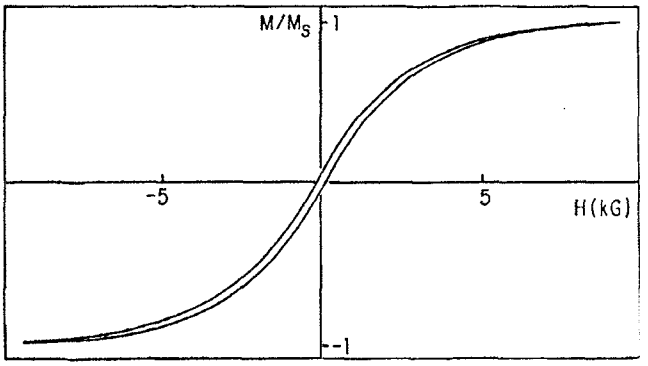

Figure 2.: Magnetization vs. in-plane field for Fe(30 Å)/Cr(12 Å) superlattice at 4.2 K (from ref. [4]).

relatively high, is characteristic of an antiferromagnet. In a ferromagnetic configuration the system saturates at low fields and the slope of M at $M \cong 0$ is much steeper. Magnetization measurements of this kind have been used to establish whether the interlayer coupling in magnetic metallic multilayers is antiferromagnetic or not. This may be confirmed by other experimental technics such as light scattering[9], SPLEED[10], neutron diffraction[11], and observations of the magneto-optical[12] and planar Hall effects[4]. Also, the magnitude of the interlayer exchange coupling in the antiferromagnetic configuration can be obtained from the observed value of the saturation field as follows[13]. The energy of two magnetic layers with magnetizations $\vec{\mu}_1$ and $\vec{\mu}_2$ coupled by exchange interaction J in the presence of a magnetic field H is

$$E = -J\hat{\mu}_1 \cdot \hat{\mu}_2 - H \cdot (\vec{\mu}_2 + \vec{\mu}_2), \qquad (1)$$

where $\hat{\mu}_1$ and $\hat{\mu}_2$ are the unit magnetization vectors. Let us consider the case where both $\vec{\mu}_1$ and $\vec{\mu}_2$ are parallel to the layers. With the in-plane field H perpendicular to the magnetization axis, the saturation field H_s is obtained by imposing that the minimum of E occurs when both $\vec{\mu}_1$ and $\vec{\mu}_2$ are parallel to H. It follows that the exchange coupling per unit area A between the magnetic layers is given by

$$J \equiv J/A = -H_s M_s t_M/2, \qquad (2)$$

where M_s and t_M are the magnetization per atom and the thickness of the magnetic layer, respectively.

The measured values of J obtained in this way are, for example, $J = -2.8$ ergs/cm^{2}in Fe/Cr(9 Å), $J = -0.24$ ergs/cm^2 in Co/Cu(95 Å), and $J \cong -5$ ergs/cm^2 in Co/Ru(35 Å). These relatively high values of J cannot be accounted for simply by magnetostatic interaction. Just for comparison, the estimated value of J between nearest neighbor bulk iron atomic planes is $\cong 18.9$ ergs/cm^2.

It has been found that the sign and magnitude of the interlayer coupling depend on the spacer layer thick-

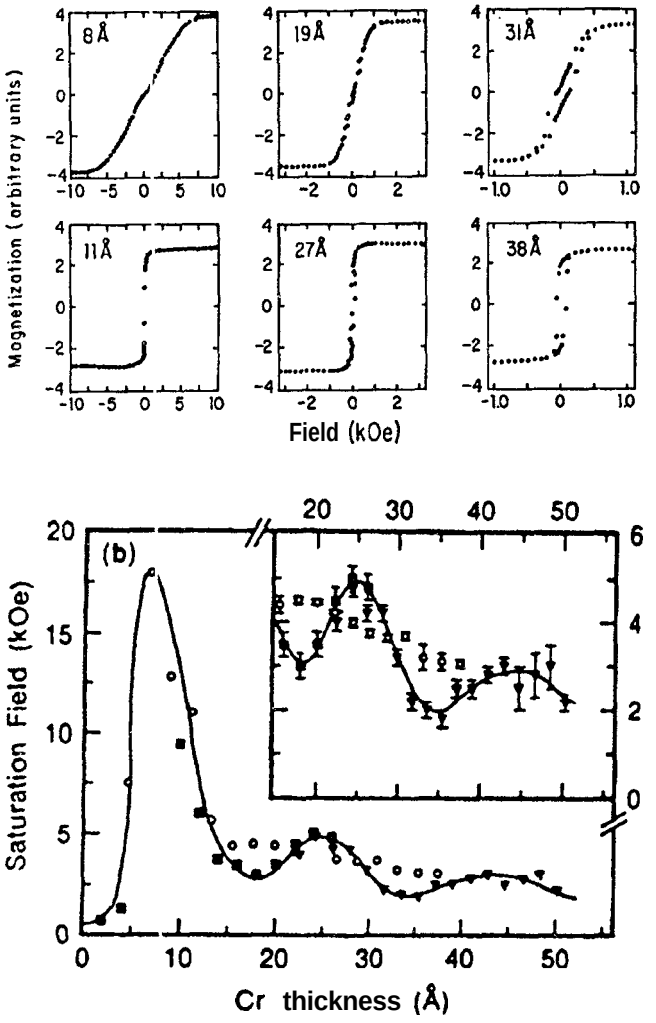

Figure 3.: (a) Magnetization (300K) vs. in-plane field for Co(18 Å)/Ru(t_{Ru}) superlattices for some values of t_{Cr} as indicated in the figures; (b) Saturation field (4.2 K) vs. Cr layer thickness for Fe(20 Å)/Cr superlattices. Taken from ref. [6].

ness. In fact, Parkin et al.[6] discovered that the interlayer coupling oscillates with an overall decreasing amplitude, as the spacer layer thickness increases (Fig. 3). The period of oscillations in sputtering grown samples can be rather large, e.g. ≅ 20 Å in Fe/Cr [6], and ≅ 12.5 Å in Co/Cu or Fe/Cu. This behavior has been confirmed by light scattering measurements[14].

More recently, the Jülich group devised a very ingenious sample in which a Cr wedge is deposited over an iron single crystal whisker and subsequently covered by an Fe overlayer. In this way, the Cr thickness is almost continuously varied, and Fe interlayer couplings through different Cr thicknesses are realized in a single sample. Both Purcell et al.[15] and Demokritov et al.[16] have observed, by magneto-optical Kerr effect (MOKE) measurements in this kind of a sample, a period of about two monolayers apparently superimposed on a long period comparable to that previously seen by Parkin et al[6]. In a beautiful experiment Unguris et al.[17] uscd scanniog electron microscopy with polarization analysis (SEMPA) to demonstrate tliat tlie interface quality plays a dccisive role as far as the period of oscillation in this system is concerned. They obtain both short and long period oscillations in the coupling by varying tlie quality of tlie wedge structure, as shown in Fig. 4. Poorer quality interfaces apparently introduce irregular local variations in spacer thickness wliich wash out short period oscillations. With flat interfaces otlier superlattices present short period oscillations superimposed to long ones[18].

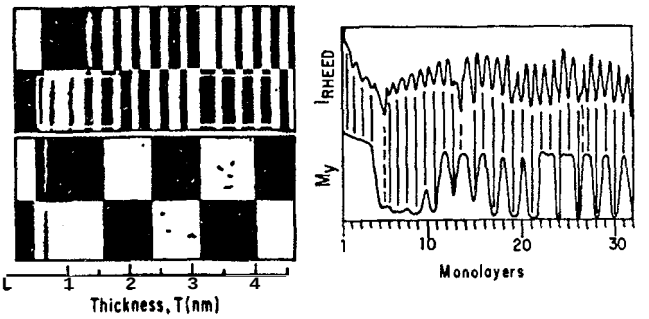

Figure 4.: (a) SEMPA images of the magnetic coupling of the Fe layers in two Fe/Cr/Fe wedge samples with different crystalline qualities. The image in tlie upper panel refers to the well ordered Cr layer spacer and shows short period oscillations in the coupling. The lower panel image refers to tlie poor quality sample and shows the same period as that obtained with sputering grown samples6, 14. (b) Magnetization and RIIEED oscillations corresponding to the good quality sample of part (a). The oscillations in M_y line up with the RIIEED. Taken from Unguris et al. (ref. [17]).

Tlie period, phase, and amplitude of the oscillations depend on the multilayer constitueiit materials and also on the crystal direction of growth[18,19]. Phase and period changes occur between the (111) and (100) directions in Co/Cu, and (100) Fe/Cu superlattices[20], as shown in Fig. 5.

IV. Electronic Transport Properties

Measurements of electronic transport properties in metallic multilayers often require the same precautions as in metallic thin films. The electrical currents must be small to avoid layer damages and the experimental geometries used must take into account the fact that total thickness of these multilayer samples are usually small.

A classical theory for the conduction in thin filrns and wires in the absence of a magnetic field was developed by Fuchs and Sondheimer[21], and later adapted to superlattices by Carcia and Suna[22]. The presence of an applied magnetic field alter the electron motion causing the conventional magnetoresistance effect. On the

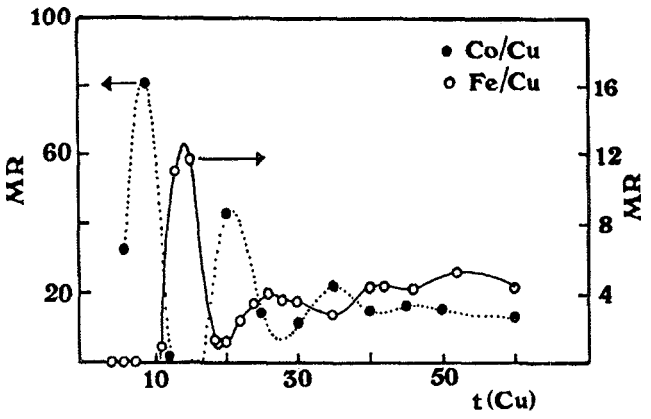

Figure 5.: Variation of the magnetoresistance ratio of both Fe(15 Å)/Cu(t_{Cu}) (open symbols) and Co(15 Å)/Cu(t_{Cu}) (full symbols) superlattices as a function of the Cu layer thickness. Dotted and full lines are just eye guides. Taken from Petroff et al. (ref. [20]).

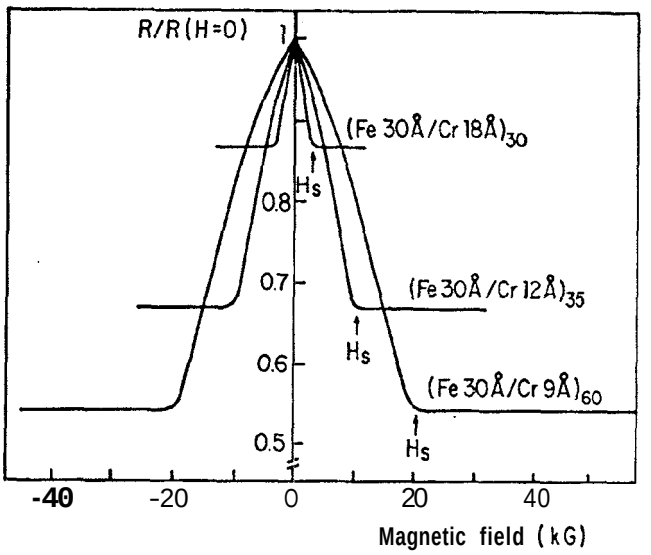

Figure 6.: Magnetoresistance of Fe/Cr superlattices at 4.2 K. Current and magnetic field are in the plane of the layers. Taken from Baibicli et al. (ref. [4]).

otlier hand, the resistivity of magnetic materials may depend on the direction of the current relative to the magnetization axis. This effect is also called magnetoresistance but, in this case, the main role of tlie field is not to influence the electron motion but merely to align the magnetic moments. The difference in resistivities for currents flowing parallel and perpendicular to the magnetization in traditional rnaterials are relatively small, not exceeding 3%. However, in some magnetic metallic multilayers a dramatic change in resistance can be produced by applying a magnetic field which alters tlie magnetic configuration of the system. A giant effect of this type was observed in Fe/Cr superlattices by Baibich et al[4]. They have studied samples grown by MBE in the (100) direction, where the magnetization of the Fe layers lie in the plane of the layers. They have measured the change in resistance as a function of applied magnetic field with the current also in the plane of the layers. No significant difference was found between tlie cases where the current is parallel or perpendicular to the field. For 9 Å Cr thickness corresponding to an antiferromagnetic coupling between the Fe layers they have observed, at low temperatures, a change of almost a factor of two between the resistivities at zero field and in the saturated stated. The same change in resistance was obtained with the applied field perpendicular to the layers, although a higher field is necessary to saturate the system, because in this case it needs to overcome both the antiferromagnetic coupling and the anisotropy which makes the magnetization to be in the plane of the layers. Typical results are shown in Fig. 6.

Giant magnetoresistance effect have been observed also in several other magnetic metallic multilayer systems[6,23]. Most of the magnetoresistance measurements in these systems are performed with the current flowing in the plane of the layers, but recently Pratt et al.[24] have shown that the magnetoresistance of Co/Ag measured with the current perpendicular to the layer planes can be more than ten times as large as the current-in-plane magnetoresistance of the same sample. Their measurements require special techniques such as SQUID detection because of tlie very small voltages and resistances involved.

The saturation magnetoresistance ratio is defined as

$$\Delta R/R = [R(H_c) - R(H_s)]/R(H_s), \qquad (3)$$

where H, is the coercive field and H, is the saturation field. It is interesting to note tliat the original definition used by the Orsay group was $[R(0) - R(H_s)]/R(0)$. Tlius, their published data for the magnetoresistance of Fe(20 Å)/Cr(12 Å) multilayer is a factor of two bigger than tliat observed by Parkin et al.[6] for the same system. The difference seems to be due to fact that they have used different samples, grown by MBE and sputtering metliods respectively. However, one would generally expect MBE samples to have sliarper interfaces than tliose prepared by sputtering, and hence less interface roughness. The role played by interfacial roughness in the magnetoresistance is not yet fully understood. Direct experimental attempts to determine whether introducing interface roughness increases or decreases $\Delta R/R$ are still inconclusive. In some cases, it seems that a certain amount of roughness is necessary to maximize the magnetoresistance effect, as schematically shown in Fig 7 [27]. But, as pointed out by Parkin (private communication), one must be careful in analyzing these experiments because small changes in the spacer thickness around a magnetoresistance maximum can place tlie sample bellow or above the magnetoresistance peak.

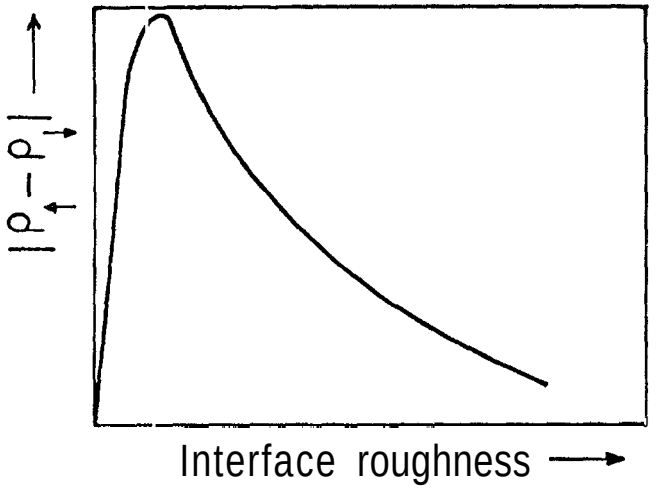

Figure 7.: Qualitative sketch of the spin dependent interface scattering diference as a function of interface rougliness. Taken from ref. [26].

Growth ccnditions may influence the quality of the samplcs introducing some dispersion in the experimental data. Nevertheless, it is undisputable that the magnetoresistance effect can be very large in some magnetic metallic multilayers. In Fig. 8 we show the results of $\Delta R/R$ in Co/Cu superlattices as a function of the Cu thickness, measured with the current in the planes of the layers. Tlie observed oscillations in $\Delta R/R$ are associated with the oscillations in the interlayer coupling. For large Cu thicknesses, the oscillations seems to disappear, and just an overall decrease in $\Delta R/R$ persists. Zhang and Levy[27] explains this behavior by arguing that when thc coupling is not sufficient strong to define a single rnagnetic configuration of the multilayer structure, it is necessary to consider an average over all possible configurations.

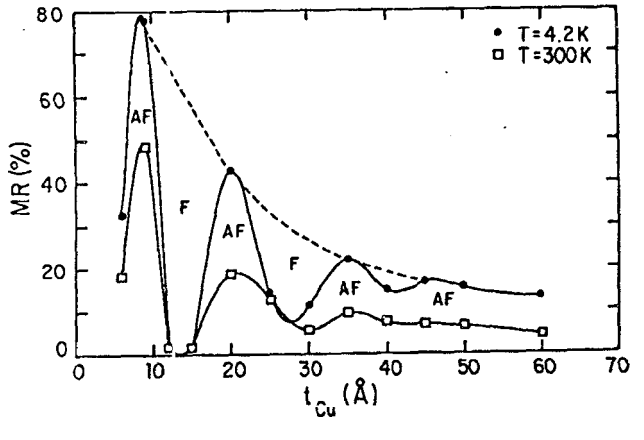

Figure 8.: Variation of the magnetoresistance ratio of Co(15 Å)/Cu(t_{Cu}) superlattices as a function of the Cu layer thickness. Taken from D. H. Mosca et al. (ref. [23]).

V.Oscillation in the exchange coupling

As we have previously mentioned, the magnetic moments of adjacent ferromagnetic layers composed by metals such as Fe and Co separated by a non-magnetic metallic layer can couple ferromagnetically or antiferromagnetically depending on the thickness of the spacer layer. Early work[5,28] indicated a monotonic decrease in the antiferromagnetic coupling between the magnetic layers with increasing thickness of the spacer layer. However, Parkin et al[6] discovered that tlie coupling in Co/Ru, Co/Cr, and Fe/Cr superlattices actually oscillates between ferromagnetic and antiferromagnetic as tlie spacer layer thickness increases. This oscillatory behavior was subsequently observed by different groups in a wide variety of other metallic systems[14,23,29]. We have also mentioned that the strength of the antiferromagnetic coupling is, in most cases, sufficiently high to exclude a possible magnetostatic origin. Also, the rather large exchange coupling period ($\simeq 10 - 20$ Å) initially observed in some multilayers caused surprise, because a naive application of a continuous and free electron gas Ruderman-Kittel-Kasuya-Yosida (RKKY) theory gives much shorter periods. These facts motivated several theoretical and experimental works aimed at understanding the origin of both the exchange coupling and its long period of oscillations in those metallic multilayers.

Generally, the exchange coupling can be determined by the difference in total energy between the ferromagnetic and the antiferromagnetic configuration of the system. Direct numerical calculations of the total energies, however, must be performed with very high accuracy because the energy differences are extremely small in comparison with the total energies involved. Hence, the degree of complexity in treating the underlying electronic structure and the method of calculation used may severely restrict the range of systems which can be practically treated by this approach. For instance, Hasegawa[30] found that the required accuracy to compute the exchange coupling is very hard to achieve even with a simplified tight-binding d-band model. First principles calculations based on local spin density functional theory have been carried out[31,32] for some superlattices but the calculated exchange coupling is one or two orders of magnitude larger than observed. Stoeffler and Gautier[33] used a multi-orbital tight-binding model to calculate the total energies of various multilayers but their results are also large when compared with the experimental values. Presently these calculations are restricted to thin layer thicknesses and hence still unsuitable to investigate long period oscillations. Model calculations with simpler band structure are important because they may reveal the relevant physical mechanisms often hidden behind extensive first-principles numerical computations. Along this line Edwards et al.[34–36] considered a simple model and developed a theory for the coupling across a transition metal spacer which exhibits

many of the observed features of tlie oscillatory intcrlaycr coupling.

Here, for presentation purposes, we distinguish between noble aiid transitioii metal spacers and divide existing tlieoretical works into two different (but in our vicw complemeiitary ratlicr tlian competing) types of approach. Theories of tlie exchange coupliiig bascd on total energy calculations like tlie ones we have briefly mentioned, aiid RKKY-type of theories in wliicli the exchange interaction between localized moments mediated by coiiduction electrons are calculated by perturbation theory[37–42].

For noble metal spacers, with tlie d bands well bcllow tlie Fermi level aiid a broad nearly-free-electron sp conduction band, it is difficult to conceive anything much different froin a RKKY-like coupliiig. As a sccond order perturbation tlieory, RKKY is not expected to provide a good dcscription of tlie coupling for thin spacer layer tliicknesses. However, in tliis case, first-principles total energy calculations has a chance of yielding accurate rcsults for the coupling. On tlie other hand, in tlie opposite limit (i.e. for very large spacer layer tliickiiesses) total energy calculations are very hard to deal witli numerically, but RKKY may provide a much simpler approacli. Nevertheless, it is difficult to calculate tlie strength of the coupling within RKKY, wliere usually tlie value of tlie coupling ultimately depends on a ratlier arbitrary choice of values for tlie parameters involved.

Asymptotic beliavior concerning tlie period of oscillation, rate of decay and tcmperature dependence obtained from RKKY[40] agree with tlie model calculatioiis of Edwards et al[34,35]. In botli theories tlie occurrence of long pcriod oscillations in tlie cxchange coupliiig is iiitiinately associated to thc discrete nature of tlie spaccr layer. For a ooe-band tight-binding model witli nearcst plane liopping, and layer orientation correspondiiig to a plane of reflectioii symmetry in tlie spacer, Edwards et al[35] showed tliat tlie period of oscillation is determined by caliper measurements of tlie spacer Fermi surface normal to tlie layer planes, and long-periods arise when tlie Fermi surface is close to tlie zone boundary. Coehoorn[43] (see also Cliappert and Renard[44]) drew similar conclusions by analyzing in real space tlie ratio between tlie period of the spin density oscillations induced in the conduction electrons and tlie discrete spacer layer thickness.

Tlie RKKY range function in a planar geometry may be calculated[45] by taking the one dimeiisional Fourier transform of the wavevector-dependent susceptibility $\chi(q = 0, q_z)$, wliere q and q_z are tlie components of tlie wave vector parallel and perpendicular to tlie layers respectively. Asymptotically, significant coiitributions to the range function mainly comes from wave vectors which maximize χ, hence long period oscillations iii tlie interlayer coupliiig are associated with singularities in $\chi(q_z)$. Bruno and Chappert[40] have analyzed bulk Fermi surfaces of Cu, Ag and Au in an extended zoiie scheme to identify tlie extrema in χ aiid determined tlie relevant wave vectors for the iiiterlayer coupling periods of oscillations in different crystalline orientations. For (111) Cu spacer layer they predictcd a unique period of r 9.4 Å in very good agreemciit with tlie value observed in Co/Cu samples predominantly textured in the (111) direction[23].

For traiisition metal spacers witli partially filled *d*-bands tliere is no reason to expect a simple nearly-frecelectron like polarization of tlie type usually considered by RKKY. A complete treatinent of tlie polarization involving tlie d-bands would be mucli more iiivolved. Wang et al[39] used a theory[46] originally developed for rare earth compounds to study the interlayer coupling in Fe/Cr. In tlieir calculations tliey have used full band structiire of bulk paramagnetic Cr but tlie ↑ and ↓ spins Fe d-bands were substituted by atomic levels locatcd below aiid well above tlie Fermi encrgy rcspectivcly. The magnitude of the coupliiig was fouiid to be strongly dependent on tlie estimated values of the position of tlie ↑ spin Fe *d*-level relative to the Fermi energy, and a second parameter was used to set tlie final coupliiig energy scale. Ferromagnetic transition mctals separated by transitioii metal spacers should have tlieir d-bands treatcd on equal footing, and preferably witliin an itinerant picture because tliey are not localized. Conseqiiently, il1 tliis case, a more careful treatinciit of tlie electron spin iiiteractions is required.

We now briefly describe tlie tlieory of Edwards et al[36] for tlie iiiterlayer coupling across a transition metal spaccr. For tliis purpose, it is sufficient to consider two semi-infinite traiisition metal fcrromagnets separated by a traiisition mctal spacer containing N atomic planes. The exchaiige coupling $J(N)$ is given by tlie difference in energy, per unit area of tlie layers, between tlie fcrromagnetic and antifcrroinagnetic configuratioiis of tlie saiidwich. For simplicity it is assumed tliat d-bands contributioiis to tlie energies are dominant aiid hence tlie *sp* conduction band is omitted; total cnergics of tlie two configurations are approximated by one-electroii energy sums. Tlie Fermi level is fixed by tlie bulk fcrromagnets aiid tlie spaccr aligned accordingly. Changes in tlie *d*-levels in eacli atomic plane witli rcspcct to tlieir appropriate bulk values are neglected, tliereby avoiding a self-consistency wliich would sliglitly cliangc tliese lcvcls near tlie interfaccs. To make tlie calculations evcn simpler we further assume tlie same d-band width for botli magnetic and non-magnetic metals, and tliat tlie number of ↓ spins electrons per atom in tlie magnctic metal is cqual to tlie nuinber of electrons pcr atom of either spins il1 tlie non-magnetic metal. To emphasize tlie basic physical mcchanism we initially consider tlie case in wliich tlie ↑ spin electron *d* band of tlie ferromagnet is full. In sucli a case, when tlie sandwicli is in tlie fcrroinagiietic configuration, thc ↑ spin Iioles experience a constant potential throughout

tlie structure, whereas ↓ spin holes feel a potential well in tlie spacer layer. The well is caused by the excliange field wliicli increases the number of ↑ spin electrons in tlie ferromagnets in both sides of tlie sandwich in tlie parallel configuration, lience reducing the ↓ spin lioles occupation n these regions. Tlie presence of the well confine the . spin lioles essentially in the non-magnetic spacer metal, and introduces size quantization effects wliicli clearly depend on tlie spacer layer thickness. When tlie sandwich is in the antiparallel configuration, ↑ spin lioles experience a potential step when crossiiig tlie second nterface, and ↓ spin lioles feels a similar stcp when crossing the first interface. It follows tliat, in tliis configuration, ↑ spins holes are confined in tlie half space tc tlie left of tlie second interface, and ↓ spin liolcs in tlie half to tlie riglit of the first interface. The situation is schematically shown in Fig. 9 for botli configurations and spin directions. The well depth, and heights of the two steps, depend on the excliange splittiiig of tlie ferromagnets and control the effectiveiiess of tlie confinements. A very simple model is to consider tlie on-site interaction $U = \infty$ in tlie magnetic layers aiid $U = 0$ in tlie spacer layer. In tliis case, ↓ spins d lioles are completely confined in the spacer layer in tlie ferromagnetic configuration, and tlie cost in energy to produce such a confinement clearly depends on tlie spacer tliickness. On tlie other liand, in tlie antiferromagnetic configuration, tlie lialf space confinement of eacli spin d holes is caused by a single surface term, and hence tlie associated cost in energy is independent of tlie spacer tliickness. Therefore, qualitatively, it is then conceivable that the energy cost to obtain a ferromagnetic coiifiguration may be sometimes higher sometimes lower than tlie antiferromagnetic one, depending on tlie spacer layer thickness.

Tlie interlayer coupling is proportional to tlie encrgy difference between the parallell and antiparallel configurations of tlie sandwich, with constant liole number, aiid is given by

$$J(N) = [\Omega(N) - \Omega(\infty)]/A \qquad (4)$$

where $\Omega(N)$ is the free energy which at zero temperature reduces to

$$\Omega(N) = E(N) - E_F n(N). \qquad (5)$$

Here, $E(N)$ s the total energy of the ↓ spins holes confined in a spacer with N atomic planes, measured relative to a reference state with N bulk planes; $n(N)$ is tlie corresponding number of lioles; E_F is the Fermi energy, $\Omega(\infty)$ is the constant (N independent) surface term present in the antiferromagnetic configuration, and A is tlie area of the layers. At zero temperature $\Omega(N)$ inay be calculatetl by

$$\Omega(N) = \int_{-\infty}^{E_F} (E - E_F)\Delta\rho(E)dE, \qquad (6)$$

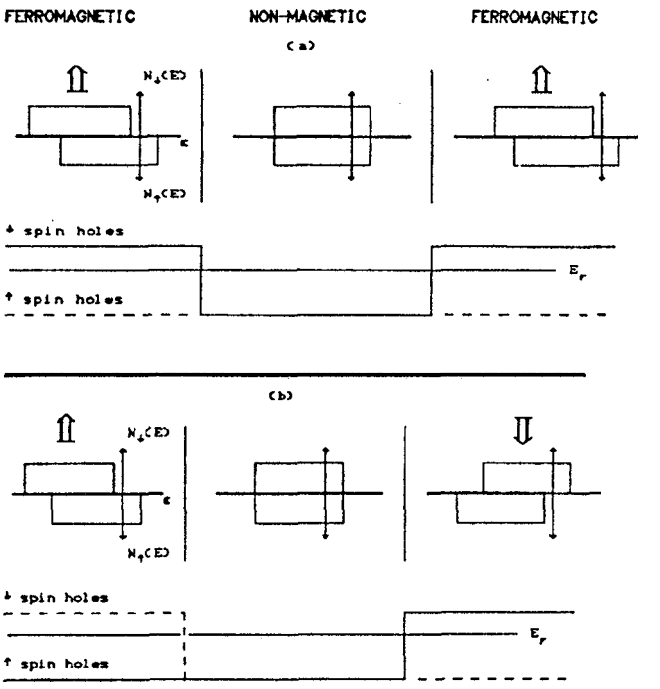

Figure 9.: Schematic representation of tlie densities of states for eacli spin $N^{\uparrow}(E)$ and $N_{\downarrow}(E)$ in eacli region of a sandwich for (a) ferromagnetic aiid (b) antiferromagnetic alignments of the magnetic layers; the vertical axis is drawn at the Fermi level E_F. Inset: Schematic plots of tlie potentials experienced by tlie lioles of different spins in eacli case (dashed line for ↓ spin lioles, solid line for ↑ spin liolcs).

where **Ap** is tlie change in tlie density of states of tlie confined holes relative to bulk reference system.

Tlie confinement quantizes tlie ↓ spin hole states in tlie direction perpendicular to the layers and this shows up as steps in tlie corresponding density of states. The number, heiglits and position of tliese steps depend on the widtli of tlie well, as in the case of a film where they depend on the film thickness. As the spacer layer thickness changes, these steps move and may cross tlie Fermi level, leading to oscillations in the exchange coupling. The situation is similar to the de Haas-van Alplien (dHvA) effect in which oscillations are associated with the quantization produced by an applied magnetic field. Here, the quantization is imposed by the excliange field and tlie variation of the spacer thickness causes tlie quantized states to pass tlirough E_F. Exploiting this analogy witli the dHvA, and considering a simple one-band tight-binding model with nearest plane hopping, and layer orientation corresponding to a plane of reflection symmetry in the spacer, Edwards et al. derived tlie following asymptotic formula for the interlayer coupling

$$J(N-1) = \frac{1}{4\pi NA}\mathrm{Re}\sum_{s=2}^{\infty}\frac{\sigma}{s^2}\left|\frac{\partial k_z^2}{\partial k_x^2}\cdot\frac{\partial k_z^2}{\partial k_y^2}\right|^{-1/2} \times \frac{\exp[2isNak_z^0(\mu)]}{T^{-1}\sinh\left[2\pi sNaT\frac{\partial k_z}{\partial \epsilon}\right]}$$

$$\sigma = \begin{cases} & \text{both 2nd derivatives} > 0 \\ \mathrm{i}\ , & \text{both derivatives} < 0 \\ 1, & \text{one derivative} > 0, \text{ the otlier} < 0. \end{cases} \quad (7)$$

The formula is valid for finite temperature T as well as T = 0, and it is exact for this particular rmodel, where $U = \infty$ in tlie magnetic metals and equal to zero in the spacer layer. Here, $k_z^0(\mu)$ is an extremal radius of the Fermi surface in the direction perpendicular to the layers (half the caliper measurement) and all the derivatives in Eq. 7 are taken at the stationary point k_z^0 r $k_z^0(\mu, k_x^0(\mu), k_y^0(\mu))$; a is the lattice parameter and μ the chemical potential fixed by the ferromagnets (at T = 0 $\mu = E_F$). The consequences of this asymptotic formula for $J(N)$ are:

(i) The period of oscillations in the exchange coupling is determined by the factor $\exp[2iNak_z^0(\mu)]$. Clearly, owing to the discrete thickness Na of tlie spacer layer, $k_z^0(\mu)$ may be replaced by $k_z^0(\mu) - \pi/a$. Therefore, long periods are obtained either when the radius $k_z^0(\mu)$ is small or when the Fermi surface approaches the zone boundary at π/a so that $k_z^0(\mu) - \pi/a$ is small.

(ii) The amplitude contains a factor of the Ferrni surface at its extrema1 points.

(iii) The temperature dependence of the oscillations is governed by the velocity of carriers at the extremal points.

(iv) The asymptotic decay at T = 0 is proportional to $1/N^2$ but becomes exponential at finite T.

Numerical calculations for specific cases of a one-band model and simple cubic (100) orientation of the layers at T = 0 show that the asymptotic formula given by Eq. 7 is rather accurate for N > 5. In this case, long period oscillations are obtained for $E_F \cong -1$, which corresponds to a situation where the Fermi surface is close to the zone boundary. For $E_F = -1.05$ the sign of J for N ≅= 5 is antiferromagnetic, and the period is ≅ 10 interatomic distances, which is close to what was observed in structures like Co/Ru[3]. Also, the magnitude of J, calculated with this value of E_F and for a spacer thickness of a few atomic planes, is ≅ 1 erg cm^{-2}, which is of the order of magnitude of the Co/Ru measured value.

For $E_F = -0.95$ the period is again long, but there is a phase shift in the oscillations compared with $E_F = -1.05$. For $E_F > -1$, the Fermi surface develops four necks in the plane parallel to the layers (equivalent to two saddle points) and the diameter of the necks is small for $E_F = -0.95$. These necks are the extrema that determine the oscillations in the exchange coupling for $-1 < E_F < 0$, and the phase shift is obtained because tlie factor σ in Eq. 7 takes a value a = 1 for a saddle point.

Short period oscillations can be obtained for example with $E_F = -2.5$. In this case the amplitude of oscillations is bigger than those with $E_F \cong -1$ because of the greater curvature of the Fermi surface at tlie extremum. In practice, roughness of the surfaces may lead to a variable effective spacer thickness which would tend to suppress short-period oscillations, sucli as those with $E_F = -2.5$, due to an averaging effect.

The theory described so far was developed assuming a complete confinement of holes. Tliis is a very stringent assumption wliich clearly should be relaxed for a weak ferromagnet like Fe which has *d*-holes in both minority and majority spin bands. In this case, lioles are not totally confined inside the spacer layer, neitlier in half spaces. To investigate the effect of partial hole confinement[36], we need to consider a finite excliange splitting V in the magnetic layers. For simplicity, we avoid a full self-consistent treatment of tlie problem and assume a simple picture in which the effective on-site energies are taken to be zero inside the spacer layer, and eitlier V or 0 inside tlie magnetic metals, depending on the configuration and spin directions of the lioles. Thus, the steps at the interfaces have height V, and basically two situations may occur. The first, wliich is pictured in Fig. 9, is when $E_F < V$ as in Co. In this case, for finite V, ↓ spin holes can penetrate a few layers across the interfaces inside the magnetic metals in the ferromagnetic configuration; the confinement will not be precisely restricted to thc spacer layer, but still will be essentially within a finite region. Similar penetrations will also happen in tlie antiferromagnetic arrangement through the corresponding finite potential step experienced by holes of either spins. The other situation is when $E_F > V$. In this case, there are hoies of both spins in tlie magnetic metals (as in Fe), and they are only partially reflected by the potential steps at the interfaces. In the ferromagnetic configuration resonances may occur in the ↓ spin holes for a fixed value of V as the spacer thickness vary.

To calculate tlie interlayer coupling at T = 0, we can still use Eqs. 4-6, but a Green function method is required to calculate $\boldsymbol{Ap}$ [36], which is then given by

$$\Delta\rho = (-1/\pi)\sum_k \mathrm{Im}\sum_n \Delta G_{nn}(E,k), \quad (8)$$

where $\Delta G_{nn}(E,\mathrm{k})$ is tlie difference between the diagonal element of the Green function in atomic plane n and the appropriate reference bulk Green function, and k is a two-dimensional wave vector parallel to the layers. For the fcrromagnetic configuration where tlie ↓ spin lioles move in a potential well of depth V we may rewrite

$$\sum_n \Delta G_{nn}(E,k) = \mathrm{Tr}[\partial \ln(1 - G^0V)/\partial E], \quad (9)$$

where G^0 is the bulk Green function.

Results for the interlayer coupling, numerically calculated using a simple cubic one-band tight-binding model for the (100) orientation of the layers, are shown in Fig. 10 for two typical cases, each corresponding to one of the two situations described above[36]. It is interesting to notice that as E_F moves above the edge of the well, the phase of the oscillations shifts by almost π, and its amplitude is very strongly reduced.

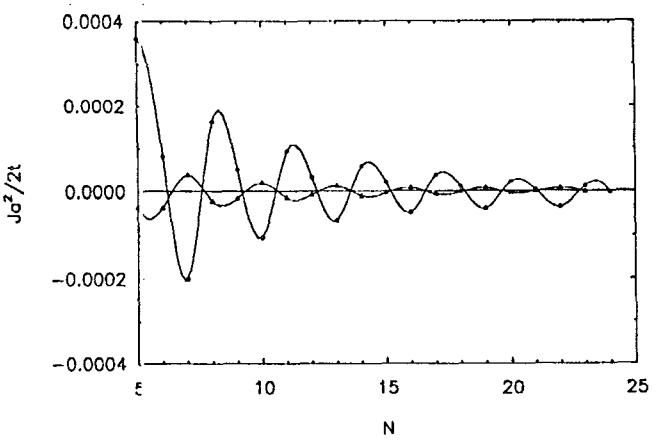

Figure 10.: Calculated values of the exchange coupling J for a simple cubic (100) one-orbital tight-binding model with nearest neighbour hopping constant t. J is plotted as a function of the number N of atomic layers in the spacer with the lattice constant a. The two curves correspond to different degrees of confinement $\epsilon = E_F/V$, where E_F is the Fermi level and V is the exchange splitting in the ferromagnetic layers. E_F is fixed at -2.5 (in units of $2t$); the circles correspond to strongly confined holes ($\epsilon = -1.0$) and triangles to weakly confined holes ($\epsilon = -6.25$).

The main conclusions for the one-band model are that the periods of the oscillation of the interlayer coupling are characteristic of the spacer metal, while the amplitude and phase of oscillations are very sensitive to the degree of confinement of the holes in the spacer, and hence depend on the matching between the spacer and the magnetic metals. Similar conclusions were obtained by Bruno[47] with a RKKY-like approach for a noble metal spacer, and experimentally, such a phase change was observed when Co and Fe are interchanged in Co/Cu, Fe/Cu multilayers[21].

Oscillations of the interlayer exchange coupling show up in magnetoresistance measurements in which the observed change in resistance is between the sample at an applied low magnetic field and at a field which saturates its magnetization. When the sample is in a ferromagnetic configuration, it saturates at low field and no significant change in resistance is seen. Hence, distinguished peaks in the magnetoresistance measurements correspond to an antiferromagnetic configuration of the multilayer.

VI. Giant magnetoresistance

The basic physical mechanism responsible for the giant magnetoresistance effect is the asymmetry in the scattering for electrons with different spins. In ferromagnetic transition metals such an asymmetry comes predominantly from the fact that the conduction electrons scatter into the d-band which has different density of final states for the two spins[48]. The existence of available d-states at the Fermi energy acts as a trap for the conduction electrons; its effectiveness being proportional to the density of available d-states[49]. In their original work[4], Baibich et al. suggested that the magnetoresistance which they have observed was coming from spin-dependent scattering at the interfaces of the multilayers. Camley and Barnás[50] worked out a detailed semi classical theory based on the Boltzmann equation, first considering only interfacial scattering but later extending it to include also spin-dependent scattering in the bulk of the ferromagnetic layers[51]. Levy et al[25] used a quantum mechanical approach based on the Kubo formalism considering both spin-dependent interfacial and bulk scattering to calculate the magnetoresistance. Both Levy et al[25] and Barnás et al[51] concluded that to explain the giant magnetoresistance effect a strong interfacial scattering is necessary. Edwards et al[52], however, showed that this is not necessarily the case and obtained bulk scattering only[53]. Whether bulk or interfacial scattering dominates probably depends on the system[54] and experimentally this question is still under investigation[55–57]. Here we shall describe the simplest theory[52] with bulk scattering only, and when the current flows parallel to the layers.

We assume an sp conduction band common to all the layers running throughout the structure. Spin-flip scattering is neglected because the mean free path associated to it is usually very large in metals. In the absence of spin-flip scattering the two spin channels are independent and carry current in parallel. A free conduction electron with spin σ travelling through the structure experiences regions with different local resistivities, because, irrespectively of what precisely causes the scattering and even if we assume a uniform density of scattering centers throughout the structure, it is scattered at a rate which is largely determined by the local density of final states at the Fermi energy. We call $\ell_{\rm min}$ and $\ell_{\rm maj}$ the mean free paths for minority and majority spin electrons in the ferromagnetic layers and ℓ_s the mean free path for both spins in the non-magnetic spacer metal. Consider for example a CoCu superlattice: in Cu the d bands are well below the Fermi energy and $\ell_{\rm Cu}$ is large. In Co, however, although the majority d band is full (which is similar to Cu), the Fermi level lies near a peak in the minority spin density of states, hence $\ell^{\rm Co}_{\rm maj} >> \ell^{\rm Co}_{\rm min}$. Therefore, the local resistivities (which are proportional to the inverse of the

local mean free paths) are different for electrons with different spins and depend on wliether the multilayer is on a ferromagnetic or antiferromagnetic configuration. In Fig. 11 the distribution of local resistivities in tlie magnetic superlattice cell is shown schematically for each spin channel and for the ferromagnetic and antiferromagnetic configurations of tlie multilayer.

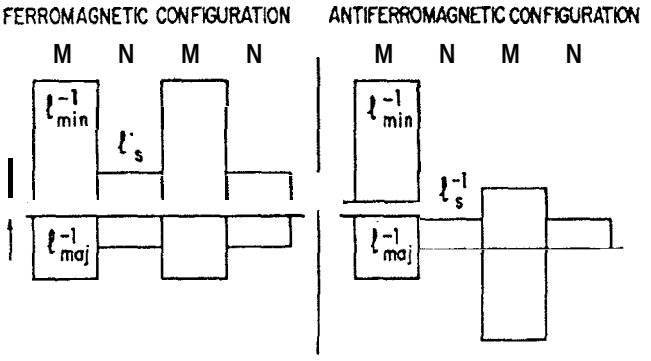

Figure 11.: Schematic representation of tlie distribution of local mean free paths ℓ^{-1} in tlie magnetic unit cell for tlie ferrornagnetic and antiferromagnetic configurations of the magnetic layers. Both the resistivities in the spin ↑ and spin ↓ cliannels are shown. M and N denote, respectively, the magnetic and nonmagnetic layers.

When the current flows parallel to the layers it is instructive to consider two extreme limits: First, when tlie longest mean free path is much sliorter than any of tlie layer thicknesses. In this case, electrons of a given spin will hardly sample different layers, which means tliat the layers will behave essentially as resistors in parallel. It is then clear that, in this case, there will be no magnetoresistance effect, i.e. $\Delta R/R = 0$, because the amount of high and low resistivities cliannels to be added in parallel will be exactly the same for both configurations of the multilayer. The second, and opposite limit, is when the mean free paths are much longer than tlie layer thicknesses. In this case, electrons of a given spin will sample many layers and will experience an average resistivity. One way of generally working out this average for superlattices is by numerically solving the Boltzmann equation with continuity and periodic boundary conditions imposed on tlie distribution function[52,53]. However, in tlie very Iong mean free path limit, a conduction electron with spin σ shall equally sample all the layers and will experience a very simple average resistivity. More specifically, in such an extreme limit, the average resistivity for each spin in tlie antiferromagnetic configuration will be proportional to

$$(M\ell^{-1}_{\rm min} + M\ell^{-1}_{\rm maj} + 2N\ell^{-1}_{s})/(2M + 2N), \qquad (10)$$

whereas in the ferromagnetic configuration the corresponding average resistivities will be proportional to

$$(M\ell^{-1}_{\rm maj} + N\ell^{-1}_{s})/(M + N), \qquad (11)$$

for ↑ spins, and

$$(M\ell^{-1}_{\rm min} + N\ell^{-1}_{s})/(M + {\rm N}), \qquad (12)$$

for ↓ spins, where M and N are tlie thicknesses of tlie magnetic and non-magnetic layers respectively. It is clear from tliese equations (or Fig. 11) tliat the lowest average resistivity occur for the ↑ spin electrons in tlie ferromagnetic configuration. This channel then acts as a shunt, making $R_{\uparrow\uparrow}$ smaller than $R_{\uparrow\downarrow}$. Also, from these straightforward averages, it follows tliat $\Delta R/R$ is given by

$$\Delta R/R = \frac{(\alpha - \beta)^2}{4(\alpha + N/M)(\beta + N/M)}, \qquad (13)$$

where $\alpha = \ell_s/\ell_{\rm min}$ and $\beta = \ell_s/\ell_{\rm maj}$. Tliis very simple analytic formula, althougli valid only in tlie litnit of very long mean free paths, helps to understand several of the magnetoresistance features observed in metallic multilayers. It is clear tliat for fixed α,β and $M\Delta R/R$ decreases as a function of N. Actually, we can see in Fig. 12 tliat Eq. 13 provides a good fit to the experimental data for reasonable values of tlie parameters a and β.

It is also clear tliat to obtain a large $\Delta R/R$ we need N/M as small as possible, C, as large as possible, and principally either α/β or β/α as large as possible. Tliis explains why CoCu with both large ratio α/β and ℓ_s is such good a combination to produce large niagnetoresistance effect. If Co is substituted by Fe, i.e. if a FeCu superlattice is considered, tlie situation is sliglitly different. The Fermi level in Fe lies in a dip of the minority spin density of states and $\ell^{\rm Fe}_{\rm maj} < \ell^{\rm Fe}_{\rm min}$ hence $(\beta/\alpha)_{\rm Fe} > 1$. However, the important thing is tliat tlie ratio $(\beta/\alpha)_{\rm Fe}$ (roughly estimated from bulk density of states calculations[58]) is srnaller than the corresponding ratio $(\alpha/\beta)_{\rm Co}$, causing the magnetoresistance of Fe/Cu to be smaller than tliat of Co/Cu as observed[**].

When irnpurities are added to the spacer metal turning it into an alloy, for a fixed M and N tlie ratio (α/β) associated to the bulk ferromagnetic metal is kept essentially constant but ℓ_s clianges. In tlie low impurity concentration limit tlie decrease in ℓ_s is proportional both to tlie impurity concentration x and to the strength of the impurity scattering potential. Therefore, $\Delta R/R$ decreases with increasing x, and the decrease is more pronounced for impurities with larger $|\Delta z|$, where $|\Delta z|$ is tlie difference between the atomic numbers of tlie two alloy components. Tliis was observed[59] in $Fe/(X_xCr_{1-x})$ superlattices for X = Ti, V, Mn (among others elements), and analyzed along these lines by Edwards et al[60]. The situation when impurities are added to the ferromagnetic metal is more subtle. It is well known that impurities in ferromagnetic transition metals can produce significant changes to their electronic structure and these changes may be very different for the two spins directions. Virtual bound states may pass through the Fermi level,

split off from the bands; screening effectiveness is usually spin dependent and may vary, among other things according to the impurity type, and as to whether the matrix is a strong or a weak ferromagnet[48,61–63]. As consequence, alloying the ferromagnet generally affect differently the density of states for electrons with different spins, and in many cases drastically changes both the magnetic and the transport properties of the ferromagnet. Fert and Campbell[64] have made an extensive and detailed investigation of the effect of diluted impurities in the transport properties of bulk ferromagnetic transition metals. They have shown that ℓ_{maj} and ℓ_{min} (hence the ratio α/β) may vary considerably according to the combination of the alloy components. The extent to which the changes in the magnetoresistance correlate to the changes in the alloy density of states as different alloy constituents are chosen for the ferromagnetic layers would be a good test to verify the importance of the present mechanism.

The simple formula on which we are basing our discussion obviously has limitations. It is valid when all the mean free paths are longer than the superlattice cell but this is not true in many cases. For example, Eq. 13 indicates that $\Delta R/R$ increases with increasing M (saturating at a value $(a-\beta)^2/4\alpha\beta$ when $M \to \infty$), but it clearly stops being valid when $h4$ is longer than the shortest mean free path. In this case, as previously mentioned, we have to solve the Boltzmann equation to work out properly the average resistivities sampled by the conduction electrons with different spins. The shorter the mean free paths, relative to the layer thicknesses, the narrower will be the region effectively sampled, which causes the magnetoresistance to decrease. Therefore, the magnetoresistance may initially increase with increasing M but it should reach a maximum and then decreases as M gets longer and we deviate further from the uniform sampling and approach its opposite limit where the magnetoresistance is zero. A simple calculation shows that large deviations from the uniform sampling limit occur for mean free paths shorter than approximately $1/4^{th}$ of the superlattice cell[52]. In a Co/Cu superlattice for example, a more refined theoretical analysis[53] of the data based on the solution of the Boltzmann equation gives $\ell_{min} \simeq 12$ Å, $\ell_{maj} \simeq 130$ Å and $\ell_s \in 260$ Å. The full solution shows that in Co(M)Cu(9 Å) superlattices $\Delta R/R$ passes through a maximum at M $\in \ell_{min}$, whereas the form of $\Delta R/R$ as a function of N does not depend much on ℓ_{min} for M $< \ell_{min}$. The maximum at $M \in 12$ Å in $Co_M Cu_9$ superlattices indicates that for M > 12 Å significant deviations from the uniform sampling becomes important, which is consistent with ℓ_{min} being less than $1/4^{th}$ of the magnetic superlattice cell. It is therefore clear that for a larger Cu spacer as in $Co_M Cu_{20}$ superlattices the maximum, if it occurs, would certainly be at a value of $M < 12$ Å (possibly at M < 4Å) as the experimental data of Mosca[65] suggests.

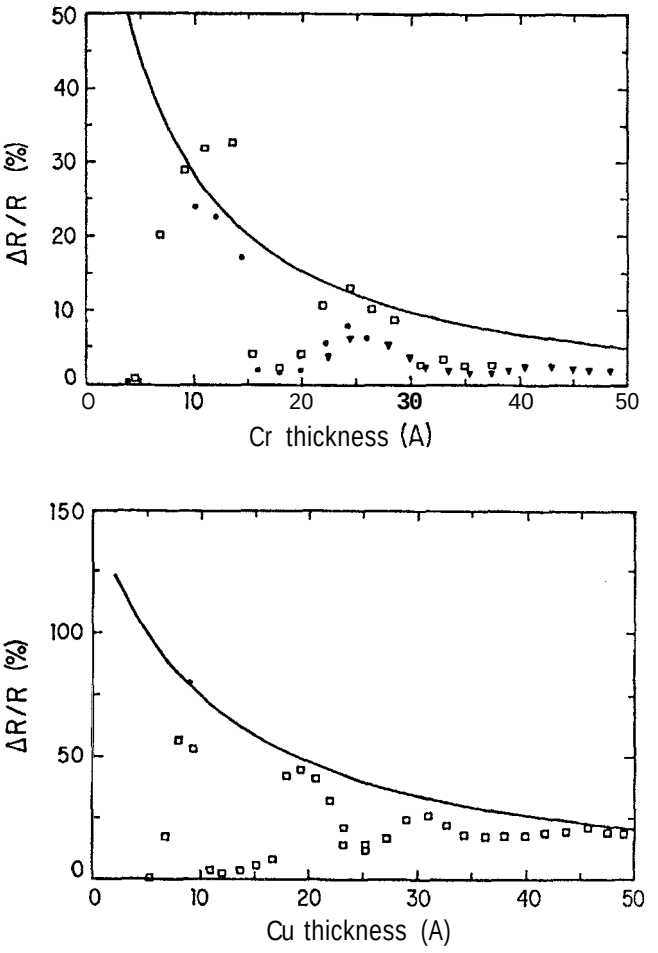

Figure 12.: (a) Dependence of $\Delta R/R$ on Cr thickness for Fe(20 Å)/Cr(t_{Cr}) superlattices. The full line is calculated by Eq. 13 with a = .4 and $\beta = 2$, and the experimental data is taken from ref. [G]. (b) Dependente of $\Delta R/R$ on Cu thickness for Co(10 Å)/Cu(t_{Cu}) superlattices. The full line is calculated by Eq. 13 with a = 10 and $\beta = 1.3$, and the experimental data is taken from from Parkin et al[23].

Let us now briefly discuss the role played by interfacial scattering in the magnetoresistance effect. Generally there will be a mismatch on the bottoms of the conduction bands of the two metals separated by an interface. In this situation, the conduction electrons would experience a potential step when crossing the interfaces. However, for interfaces between two transition metals or between transition and a noble metals these steps are small compared to the Fermi energy hence, as a reasonable approximation, they may be neglected. Nevertheless, when the interface is not sharp the two metals mix and in the most simple picture will form an alloy. As we previously mentioned, alloying a ferromagnet may significantly alter ℓ_{min} and ℓ_{maj} hence the asymmetry ratio α/β. Other effects due to lattice spacing mismatch between the two metals, existence of terraces, structural defects etc... may also contribute

to make the asymmetry in scattering at the interfaces be very different from its corresponding bulk value. A transport theory including a detailed description of tlie possible inhomogeneities at tlie interfaces would certainly be too laborious. A very simple approach is to treat the interface as a third material in whicli the associated mean free paths ℓ_{min} and ℓ_{maj} are considered as arbitrary parameters. Such treatinent ignores variations in tlie composition witliin tlie interface widtli, wliich is reasonable when both mean free paths are long on the scale whcre sucli variations occur. In tliis case, tlie Boltzmann equation nceds to be solved for a eight-componcnt system instead of four when interface scattering is neglected. Within tliis straightforward generalization, tlie two additional parameters ℓ^{i}_{min} aiid ℓ^{i}_{maj} associated with tlie interfaces may also be determined so as to fit experimental data, and, in this way, tlie relative iinportance between bulk and interface scattering could be estimated for a particular systern. We should bear in mind tliat tlie magnctoresistance effect depends on tlie widtli of tlie region effectively sampled by tlie conduction electrons. To obtain tlie effcct it is essential tliat at least two magnetic layers are samplcd (from Fig. 11 we can clearly see tliat if pairs of magnetic/nonmagnetic layers are treated as resistors in parallel there will be no magnetoresistance effcct). Tlierefore, thc relative importance between bulk and interfacial scattering iii $\Delta R/R$ may also depend on tlie relation between tlie mean free paths and the layer tliicknesses, because in some cases it may happen that only a fraction of tlie inagnetic layers are effectivcly sampled. Clearly, better experimental characterization and control of interface quality will be decisive on establishing whether bulk or interface scattering dominates in eacli case. Reliable theoretical calculations of tlie electronic structures are also very important to understaiid tlie clianges which occur at tlie interfaces and liow tliey correlate to tlie assymmetry in tlie scattering of clectrons witli different spiiis.

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