Theory of exchange coupling in magnetic multilayers

by B. A. Jones

Two layers of magnetic materials separated by a nonmagnetic "spacer" layer display an exchange coupling between the separated magnetic layers, as detailed by the papers on experimental aspects of magnetic multilayers in this issue. Here we describe from a personal perspective the current theoretical understanding of exchange coupling in magnetic multilayers. The understanding by this point is quite good, and involves contributions from a number of authors, of which we review a key subset. The organization of the paper is as follows. After an introduction to RKKY coupling, the next section discusses the effects of a realistic band structure (with emphasis on the work of Herman et al., Bruno and Chappert, and Stiles). The following section, which introduces quantum wells, covers the work of Mathon et al., Bruno et al., and this author. Finally, we include a discussion of the predictability of amplitude, period, and phase, including the effects of disorder.

Introduction

The theory of exchange coupling in magnetic multilayers has its basis in the interactions found between two magnetic impurities in a metal. Taking the simplest case of spin-1/2 (and avoiding issues of crystal electric fields and spin-orbit coupling), hybridization between the s–p conduction electrons of the host metal and the d- (or f-) electrons of the magnetic impurity produces an effective on-site exchange coupling at the impurity site. For s–p/d hybridization, the sign of the interaction is typically antiferromagnetic, as the conduction electrons attempt to screen the spin of the impurity in their midst. Rather than forming a negative spin-1/2 at the impurity site, however, the electrons instead spin-polarize in concentric rings around the impurity. The source of the rings of alternating polarization is that a true delta-function in space would require, in Fourier k-space, all the k-vectors from 0 to infinity to be equally weighted, viz., $\delta(r) \propto \int_{-\infty}^{\infty} dk\, e^{ikr} \cdot 1$. Since the host is a metal, there are, however, k-vectors only from 0 to the Fermi wave vector. The system thus cannot form a localized screening of the impurity spin, but does the closest alternative possibility, which results in an opposite alignment close in which overscreens the impurity, followed by a parallel alignment further away which overcompensates in the opposite direction, and so on with decreasing amplitude out to infinity. This is shown in **Figure 1**.

The periodicity of the alternations of polarization is λ_F, where λ_F is π divided by a characteristic wave vector of the Fermi surface. For the simple case of a free-electron band structure, there is only one wave vector, the Fermi

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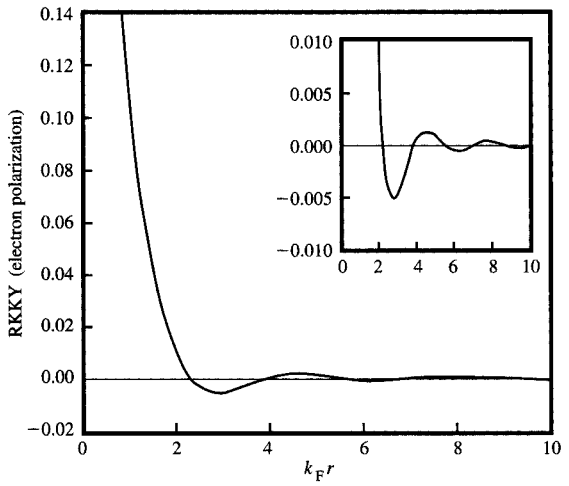

Figure 1

RKKY coupling between two spin-1/2 magnetic moments separated by a distance r, in three dimensions. Alternatively, the figure can be viewed as the negative of the electron spin polarization as a function of distance around a single magnetic moment. A positive value corresponds to ferromagnetic RKKY coupling or down-spin electron polarization, with unspecified units for both. Note that the value at $k_F r = 0$ is sensitive to the band edges, and diverges in three dimensions with increasing bandwidth. The distance r is scaled by the Fermi wave vector. Inset: the same plot, with expanded vertical scale.

wave vector of order 1 $Å^{-1}$, and λ_F is around 3 Å. Thus, within distances of 3 Å from the impurity, the conduction electrons are aligned antiferromagnetically with the impurity, from 3 to 6 Å ferromagnetically, and so on. The amplitude decay goes as $1/r^3$ for three dimensions, and varies with the geometry. Such oscillations in spin polarization have been seen experimentally [1].

Interactions between two impurities arise when they are close enough to have appreciable overlap of their oscillatory screening polarizations. If a second impurity is within the spin-down region of the first impurity's conduction electron polarization, it is favored to point oppositely to the polarization, and thus ferromagnetically with the first impurity. If the second impurity is further away, in the ring of spin-up, the second impurity prefers to point down, or antiferromagnetically with the first impurity. Thus, there is an interaction between two impurities induced by the spin-polarized conduction electrons with which they interact. The interaction takes the same distance dependence as the spin polarization, but is of opposite sign. This interaction of magnetic spins mediated by conduction electrons is designated as the RKKY interaction, after Ruderman, Kittel, Kasuya, and Yoshida [2], who independently discovered it. A typical RKKY plot for a free-electron conduction band is illustrated by Figure 1.

We can express the interaction of two spin-1/2 impurities in a sea of conduction electrons in a simple Hamiltonian form:

$$H = \sum_{\mathbf{k}} \varepsilon_k n_{\mathbf{k}} + J[\vec{s}_c(\vec{r}_1) \cdot \vec{S}_1 + \vec{s}_c(\vec{r}_2) \cdot \vec{S}_2]. \quad (1)$$

In second-quantized notation, for example, $\eta_{\mathbf{k}} = \Sigma_{\mu,\mathbf{k}} c^{\dagger}_{\mathbf{k}\mu} c_{\mathbf{k}\mu}$ and

$$\vec{s}_c(\vec{r}_1) = \sum_{\mu\mu'\mathbf{k}\mathbf{k}'} e^{i(\mathbf{k}-\mathbf{k}')\cdot\vec{r}_1} c^{\dagger}_{\mathbf{k}\mu} \frac{\vec{\sigma}_{\mu\mu'}}{2} c_{\mathbf{k}'\mu'}, \quad (2)$$

with $\vec{\sigma}$ with Pauli spin matrices. Taking the exchange coupling J to be small compared to the Fermi energy characteristic of the first term, one can perform a perturbation expansion in J/ε_F. The leading-order term is of the form $-J^2 \vec{S}_1 \cdot \vec{S}_2 f(r)$, where $f(r) = f[2k_F|\vec{r}_1 - \vec{r}_2|]$ has the form of Figure 1, and for free electrons in three dimensions can be written as

$$f(x) \propto \frac{\sin(x) - x\cos(x)}{x^4}. \quad (3)$$

Of course, Equation (3) is just a perturbation expression, and the first-order term at that. Sometimes Equation (3) is cited as the "RKKY interaction," but this is incorrect. The RKKY interaction is the exchange coupling which arises between two (or more) spins interacting via spin polarization of conduction electrons—whether J be small or large, perturbative or not, and summed to all orders. In the cases when J is small, however, the exchange interaction calculated according to the prescription above is a good approximation.

For magnetic atoms in layers, the physical interaction arises from both the interfacial and the bulk magnetic atoms interacting with the conduction electrons in which they are immersed. The interactions between each magnetic spin and all other atoms must be superposed. Those at the interfaces have the strongest effect, both because they are closest to the other layer ($1/r^3$ effect, which when integrated over a layer becomes $1/r^2$) and because they are more immersed in the spin sea of the spacer conduction electrons. For a rougher interface, there is even more mixing of magnetic and nonmagnetic atoms. Because of the presumed translational symmetry of the system in directions parallel to the layers, the interaction sum between each magnetic atom and all other magnetic atoms reduces to the sum along a line within one layer of interactions with all other atoms. A further simplification is typically achieved by assuming no spin-flip: that each magnetic moment is ferromagnetically aligned with every other atom in the same layer. Sums of interactions within

a magnetic layer are thus avoided. Yet a further simplification is to assume, rather than discrete spins, a continuous constant spin density within each layer. The sums over the second magnetic layer thus become integrals.

Either integrals or discrete sums of the system with a spherical Fermi surface give a total RKKY interaction for multilayers with many of the characteristics of impurities in bulk: The exchange interaction between layers oscillates as a function of distance between the layers, with a periodicity $\lambda_F = \pi/k_F$. The amplitude of the interaction falls off as a function of distance, but with a more complicated envelope than $1/r^3$—roughly $1/r^2 - 1/r^3$ [3]. And the physical basis of the interaction is still the spin-polarization of the intervening conduction electrons.

In comparison to real multilayer systems, of course, the most salient disagreement with the above description, discovered early on, is the difference in periodicity. As discussed in other papers in this issue, typical periodicities are closer to 11 Å than 3 Å, a discrepancy that led to the enhancements in the theory discussed in the following sections.

Realistic band structure

From the previous section we conclude that the periodicity of exchange coupling oscillation depends on wave vectors of the Fermi surface. Since the Fermi surface of real materials is not a sphere, it became clear that an accurate period must include realistic band-structure effects. Two notable approaches used were those of Herman (with Van Shilfgaarde) and of Bruno and Chappert and later Stiles, described below. (We note once again that in this paper we are reviewing primarily current understanding, not a full history of the theory. Many more theorists could be included in this and other sections; however, as ways in which band-structure tools were used to enhance our understanding, the works below are, in the opinion of this author, exemplary.)

F. Herman Herman and his collaborators, Van Shilfgaard and, earlier, Sticht, performed state-of-the-art, (nearly) self-consistent, density-functional total energy calculations [4–6]. These calculations involve setting up a unit cell which is then periodically repeated to infinity. (The systems are thus true multilayers, rather than sandwiches.) Because of the large number of states involved, the magnetic layers are seldom taken to be more than two or three atomic layers thick (compared to the 20 layers and more of experiment). This thickness is sufficient to form a magnetic moment in the layer. Spacer layers are incremented one atomic layer at a time in order to model increasing spacer width. For a given spacer width, the magnetic layers are first all aligned ferromagnetically, and the energy levels of the system are calculated and then summed over all occupied states, making it possible to obtain the total energy of the ferromagnetic system ($E_{TOT,F}$). Next, for the antiferromagnetic configuration, every other magnetic layer is fixed to lie in the opposite direction, and the total energy again calculated ($E_{TOT,AF}$). Subtracting the second total energy from the first ($E_{TOT,F} - E_{TOT,AF}$) gives a measure of the exchange energy. A positive value indicates ferromagnetic coupling preferred, and a negative, antiferromagnetic.

As can be imagined, the number of states for each system is very large, and each sum $E_{TOT,F}$ or $E_{TOT,AF}$ is an extremely large number, compared to their difference. The accuracy of the subtraction must be done to one part in 10^6 or better. Nonetheless, Herman and collaborators obtained interesting results [5, 6]. What they found was an oscillating exchange, in which the sign of the coupling alternated for every additional spacer layer. The ostensible periodicity was thus a lattice spacing, or around the familiar 3 Å once more. However, the amplitude envelope did not decay in a monotonic fashion, and upon fitting their numerical results to *two* oscillations with different amplitudes, periods, and phases, they found a good fit. One period, the dominant one, was small, 2–3 Å. The other period, with smaller amplitude, was quite large, 12–15 Å or more, depending on the material and crystallographic orientation.

It was not until several years later that experiments at NIST [7] and elsewhere showed that a second periodicity did occur in many real samples as well, with one period short and the other period long. The better the interface quality, the larger the amplitude of the short-range oscillation, which is not seen in samples where the surface roughness averages over any small-period oscillation.

The work by Herman et al. is valuable in that it pointed out the strong materials dependence of the coupling period. The work was also among the first to calculate different oscillation periods for different crystallographic orientations in the same material. Another valuable contribution involved subsequent calculations on alloys, in which the band structure of the alloy was determined by continuously interpolating between the band structures of the two constituent elements, with weighing proportional to the relative percentage of each element in the alloy. Exchange coupling periods thus calculated bore a strong correspondence to those measured by Parkin et al. in the same systems [4].

P. Bruno and C. Chappert Concurrently with the work of Herman et al., Bruno and Chappert [8] undertook a generalized analytic approach with an explicitly RKKY formalism. They also used features of the Fermi surface in various crystallographic orientations to predict long periods and multiple periods. Their use of experimentally determined Fermi surfaces enabled them to make explicit

predictions for Cu, Ag, and Au spacer layers, which were found (for Ag and Au) to agree very closely with later measured experimental periods [7] (agreement even closer than that obtained by theoretically calculated Fermi surfaces). The strong agreement provided, at an early stage, convincing indications that the RKKY interaction was the correct interaction for these systems. Bruno and Chappert also made useful remarks about the role of roughness to filter out the short periods, and about the difficulty of correct determination of the phase. Their work has had considerable historical impact; the methodology was clearly laid out, enabling other researchers to complete important extensions.

M. Stiles Stiles provided considerable depth to the Fermi surface analysis approach [9], and many of his ideas and calculational data form the basis of current thinking about Fermi surface contributions. It was not clear from the total energy and previous analytic calculations which features of the Fermi surface were giving rise to which periods (if indeed it was the vectors of the Fermi surface that gave the longer periods). Stiles performed state-of-the-art calculations of the Fermi surfaces of a wide range of spacer materials. In separate analysis, he found the conditions for the key k-vectors responsible for the exchange coupling; then, for every Fermi surface he elucidated the particular k-vectors that would be relevant for that material. For every crystal direction, there are typically a large number of candidate k-vectors k_i and, hence, periods π/k_i; to give some idea of the predominance of some over the others, he weighted the period by the degree of representation of its extremal orbit on the Fermi surface. Still, this often gave a prediction of many periodicities for each crystal direction, in contradiction to experimental observations of one or at most two. To obtain the exchange coupling exactly necessitates integrals of matrix elements of the wave functions, quantities that are difficult to obtain accurately. Variants of the Stiles approach were used by other authors, as is shown below.

Quantum wells

The geometrical effects of finite-thickness layers have one result in addition to the above RKKY description, namely quantum wells. Theories postulating variants of quantum wells in magnetic multilayers appeared fairly early (see discussion of theories below), but the definitive experimental proof came with the inverse photoemission measurements of F. Himpsel [10]. He measured the photoemission intensity of Cu on Co as a function of Cu thickness, and observed oscillations every few additional Cu layers. He ascribed the successive peaks to the formation of new bound states in the quantum well that formed in the copper. Other groups soon followed, with evidence for quantum wells in Fe/Au, Fe/Ag, Co/Cu, and other systems [11]. As proposed by Jones and Hanna [12] and others, the source of the wells was a misalignment in the bottom of the conduction bands of two adjacent layers of dissimilar materials when the chemical potentials were aligned. Of course, this means that not all systems have wells in the spacer—some have wells in the magnetic layer, and some have wells for one spin direction and barriers for another. However, quantum wells are now an accepted fact in magnetic multilayers, and it is of interest to discuss what effect they have on the exchange coupling.

One matter that is clear is that quantum wells, as a geometrical feature alone, cannot be the sole source of exchange coupling. The proof of this is the exchange coupling observed between two separated magnetic impurities in a nonmagnetic metal, where the geometry clearly does not support a well (and the bound states, if any, at each delta-function scatterer play no essential role in the exchange coupling). Moreover, dissimilar nonmagnetic metals placed in juxtaposition likewise have a mismatch of bands, but no coupling between nonmagnetic metals arises. Exchange coupling between separated magnetic layers occurs because of the spin polarization of intervening conduction electrons, and associated magnetic scattering of conduction electrons with the moments in the magnetic layer. Quantum wells cause bound states in addition, and the focus of the theories below is on the effect of quantum wells on the oscillation period, amplitude, and phase.

B. Jones and C. Hanna Although Mathon et al. published work [13] on infinitely high quantum wells at an early stage, Jones and Hanna [12, 14] were among the first to consider quantum wells of a finite height applied to magnetic sandwiches. Their mechanism, of a mismatch of electronic structure between two materials placed in contact, is currently the accepted explanation of quantum wells in magnetic multilayers. Moreover, they showed how a new feature such as quantum wells can be straightforwardly incorporated into a traditional RKKY calculation. The first-order term in an RKKY calculation is the perturbative effect of the exchange coupling on the nonmagnetic Hamiltonian. The leading term, which is of second order in the exchange coupling J, has the form

$$\Delta E = -\left(\frac{J}{n}\right)^2 \sum_{i,j} f(\mathbf{r}_i, \mathbf{r}_j)\vec{S}_i \cdot \vec{S}_j , \tag{4}$$

where $(J/n)^2 f(\mathbf{r}_i, \mathbf{r}_j)$ is termed the RKKY interaction between spins i and j, and n is the density of conduction electrons. Denoting the conduction-electron wave functions that diagonalize the unperturbed Hamiltonian by $\psi_{\mathbf{k}}(\mathbf{r})$, one obtains

$$f(\mathbf{r}_i, \mathbf{r}_j) = \frac{1}{2} \sum_{\substack{\mathbf{k},\mathbf{k}' \\ \varepsilon_k > \varepsilon_F \\ \varepsilon_{k'} < \varepsilon_F}} \frac{\{\psi^*_{\mathbf{k}'}(\mathbf{r}_i)\psi_{\mathbf{k}}(\mathbf{r}_i)\psi^*_{\mathbf{k}}(\mathbf{r}_j)\psi_{\mathbf{k}'}(\mathbf{r}_j) + h.c.\}}{\varepsilon_k - \varepsilon_{k'}}. \quad (5)$$

Logarithmic contributions at those wave vectors such that $\varepsilon_k - \varepsilon_{k'} = \varepsilon_F$ give oscillations with wave vector(s) $q_i = 2k_{F,i}$. Thus, from Equation (5), a knowledge of the wave functions and energies of the "unperturbed" (nonmagnetic) part of the Hamiltonian is sufficient to give a good estimate of the RKKY coupling.

For a quantum-well system, the wave functions $\psi_{\mathbf{k}}(\mathbf{r})$ are straightforward to calculate exactly for free-electron bands. The eigenstates are products $\psi_x\psi_y\psi_z$. Parallel to the interfaces, ψ_x and ψ_y are plane waves. Perpendicular to the interfaces, ψ_z is oscillatory for $\hbar^2k_z^2/2m$ greater than the height of the well, with a wave vector k_z that is discontinuous in space and forms a continuum in energy. For perpendicular momentum which is not large enough to escape the well, the eigenstates are bound, and decay exponentially outside the well. Their energy spectra are discrete. Although technically all states of the system are quantum-well states, since they form an orthonormal set in a quantum-well system, it is typically the bound states that are given greatest focus, since they are not present in the absence of a well. The bound states, although finite in number, contribute to the exchange coupling with a strength comparable to that of the continuum states, because the wave functions are normalized essentially to the width of the well, and therefore have a finite magnitude. The continuum states, meanwhile, although much greater in number, are each normalized to the system size, and thus the amplitude of an individual eigenstate goes to zero in the thermodynamic limit.

The quantum-well states, bound and continuous, have a number of effects on the exchange coupling. First of all, there is a phase shift imposed, with an effective delay in the well with the introduction of bound states. Second, the amplitude can be enhanced over RKKY coupling without quantum wells. If quantum wells are chosen on the basis of the band structure of the constituent materials, the quantum-well period ends up being the same as that derived from analysis of the Fermi surface, however. Thus, the main effect of quantum wells is primarily to enhance the amplitude of observable oscillations, but also to make the initial phase more indeterminate. (The effect of disorder is discussed further at the end of this paper.) An example of the effects of a relatively small quantum well is shown in **Figure 2**.

G. Mathon and collaborators The theory of Mathon and collaborators has evolved over several years [13, 15, 16]. Its current, and most comprehensive, form is as follows [16]. Quantum wells are formed by the exchange

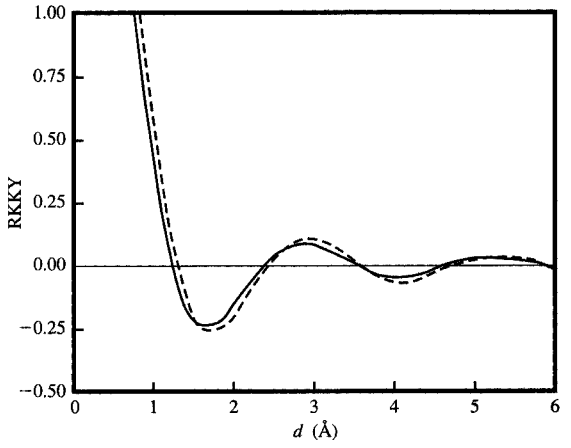

Figure 2

RKKY coupling between two magnetic layers separated by a distance d, for free electron bands. The units for RKKY coupling are unspecified. The Fermi energy is taken to be that for Cu. Solid line: Free electrons, no potential well. Dashed line: Effect of a small potential well relative to the Fermi energy. A phase shift is induced, and the RKKY coupling is enhanced for separations d greater than about 0.5 Å.

potentials of the magnetic layers, which spin-split the bands, and are an independent potential regardless of the orientation of the opposite layer, a view in accord with most current thinking. Exchange coupling between two magnetic layers is expressed in terms of thermodynamic potentials, which are themselves a weighted integral of spectral density. The net are sums over energy and $k_\parallel$, which would need many k-points for convergence. Arguing, as Stiles [9] and others have, that oscillation periods π/k_i arise from extremal radii k_i of the bulk spacer Fermi surface in the direction perpendicular to the layers, a stationary phase method is used to evaluate the sums at just those extremal k-points (typically "bellies" and "necks"). Thus, the possible oscillation periods are determined by the same Fermi surface analysis as, for example, the work by Stiles. However, this method also obtains amplitudes. To obtain the weighting for each k_i-point, the spectral density is calculated. It is found to be broad and oscillatory for some k_i, narrow and high around others. Broad, low resonances give small contribution to the exchange coupling, while delta-function-like contributions determine the observable exchange periods. Narrow delta-function-like spectral densities are just those arising from tightly confined quantum well states, so it is the k-directions corresponding to quantum wells which give the dominant periodicities. Mathon et al. make the additional argument that relying on monolayer spacers for

estimates of predominant periods is misleading, because a full quantum well forms only from bulk-like band structures of several layers or more. Mathon et al. also have a so-called torque method, which involves full converged sums over k-states, and which has good agreement with the method described above.

P. Bruno Bruno recasts the equations of exchange coupling in the presence of quantum wells in terms of reflection and transmission coefficients [17, 18] (a technique Stiles also used), and by exchanging the order of integration, obtains some analytic expressions. This method of calculation is in fact exactly equivalent to the perturbative method of Jones and Hanna (since reflection coefficients obviously depend on properties of the eigenstates). Rather than a physical explanation in terms of spin-polarized conduction electrons in the spacer, the reflection coefficient methodology can be interpreted in terms of a quantum interference mechanism. The standing waves of RKKY can of course be re-expressed as a superposition of traveling waves, each reflecting off one interface. The methodology of Bruno et al. simplifies the calculations and allows the calculation of oscillation dependence on ferromagnetic thickness and on overlayer thickness as well. Exchange coupling with varying ferromagnetic layer thickness results in an oscillation period which depends on the ferromagnetic Fermi surface. Varying overlayer thickness results in weak oscillations with the periodicity of the overlayer material [15]. The facts that the eigenfunctions of the whole system depend necessarily on the Fermi surface of each constituent layer, and that any sum over a finite number of k-vectors gives oscillations that reflect extremal features of the Fermi surface(s), make it straightforward to understand that varying the width of one layer or another merely explores that part of a larger phase space. Mathon and collaborators [19] have also obtained similar results.

Effects of disorder

Real interfaces are of course not atomically smooth, and the question naturally arises as to the effect of an uneven interface on the various calculations discussed above. If the exchange coupling oscillates with spacer width, a variable spacer distance tends to average over oscillation periods on either side of the average. For a short oscillation period, this tends to average it out entirely for a large enough amount of roughness. The longer periods of 10 Å and more are much less affected. The effects of roughness in real systems thus explain two discrepancies of total energy calculations such as those of Herman et al. [6], which showed that shorter-period oscillations have much larger amplitude than do longer-period oscillations (in contradiction to many experiments) and which gave an amplitude nearly an order of magnitude larger than experiment overall. Roughness averaging in real materials reduces the amplitude of all periods, and reduces the amplitude of short-period oscillations much more than that of long-period oscillations. Since averaging is done over spacer widths both longer and shorter than the average, the actual period remains unchanged. Calculations of delta function scatterers at the interface by Jones and Hanna [12] showed that interface scattering caused instead an additional phase shift, in addition to any quantum well phase shift present.

Exchange coupling in real systems

Finally we come to the question of theoretical predictability of exchange coupling in real systems. The best-known quantities are the oscillation periods, which can be predicted with ever-increasing accuracy for a range of materials, including alloy spacers, by examination of the Fermi surface. The fall-off with distance of the envelope of the oscillation is also well understood. The predominance of quantum wells in exchange-coupled systems is well established. The amplitude is less clearly predictable, since it decreases with increasing interface roughness and changes with the existence of quantum wells. However, empirical studies do well here, and samples of similar quality usually have similar amplitudes, with the amplitude between different systems varying within fairly well-known bounds. Moreover, as sample quality increases with time, there is a slow but steady convergence toward calculated amplitudes. Least predictable is the phase of the oscillation, since there is fairly strong dependence on disorder, although models of interface disorder by various groups are giving increasingly better fits even here.

Concluding remarks

Exchange coupling in magnetic multilayers is at this point a fairly well-settled field, with many strong contributions in the literature. It is hoped that this review of some of the key theoretical contributions will give an indication of the range of theoretical understanding achieved in this field.

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Received January 13, 1997; accepted for publication May 21, 1997

Barbara A. Jones *IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120 (BAJONES at ALMADEN, bajones@almaden.ibm.com).* Dr. Jones is a Research Staff Member and Manager of Magnetic Materials and Phenomena in the Science and Technology function at the Almaden Research Center. She received an A.B. degree in physics from Harvard University in 1982, and M.S. and Ph.D. degrees in physics from Cornell University in 1985 and 1988, respectively. After postdoctoral research at Harvard University, she joined IBM at the Almaden Research Center in 1989. She has worked on theories of magnetic impurities in metals and on superconductor surfaces, and of quantum wells and other effects in magnetic multilayers. Dr. Jones is a member of the American Physical Society; she is currently an elected member-at-large of the Executive Committee of the Division of Condensed Matter Physics.

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