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ORIGIN OF THE MAGNETIC PROXIMITY EFFECT

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Abstract

The magnetic proximity effect (MPE) has attracted the attention of theorists and experimentalists for at least three decades. Lately, the relevance of the effect for the development of nanodevices has revived interest on the subject. Here we review how the field has evolved, centering our attention on metal-metal and metal-insulator systems. We describe, and critically compare, the different theoretical approaches that have been put forward, as well as their limitations. An evaluation of the relationship between existing theories and available experimental results is also attempted.

1. INTRODUCTION

Almost 30 years ago Zuckermann [1], in a pioneer work, showed theoretically that a system formed by a thin film of a weak itinerant ferromagnet (FM), in atomic contact with a thick film of an enhanced paramagnetic metal (PM), can exhibit a nonzero Curie temperature. Shortly thereafter this theory was extended to a system formed by a ferromagnet in contact with an antiferromagnet [2]. The technique that was used consisted in solving a simplified version of the integral equation derived from the Landau-Ginzburg theory of phase transitions. This equation reads as follows:

$$M(\vec{r}) = \int d^3r' \; U(\vec{r}) \; \chi_T(\vec{r} - \vec{r}'; 0) \; M(\vec{r}') - U(\vec{r}) \; M^3(\vec{r}) \; \sum_{\omega} G_p^0(\omega) \; , \tag{1.1}$$

where $M(\vec{r})$ is the local magnetization at point $\vec{r}$, $U(\vec{r})$ the Hubbard exchange constant, $\chi_T(\vec{r}; 0)$ the static $\vec{r}$-dependent magnetic susceptibility of the non-interacting conduction electrons and $G_p^0(\omega)$ the propagator for these momentum p conduction electrons.

The above equation was analyzed using the procedure set forth by Werthamer [3], for the proximity effect between two superconductors. This way the integral equation 1.1 is transformed into a differential equation. The solution of this equation was obtained by matching the superconducting order parameter and its slope at the interface. However, this procedure is not reliable when applied to magnetic interfaces. First, with the possible exception of very weak magnets, the coherence length of the magnetic order parameter $M(\vec{r})$ is smaller than a typical lattice parameter, which excludes a continuous differential equation as a valid description. In addition, there is no *a priori* reason for requiring that the slope of $M(\vec{r})$ be continuous across an interface. For all these reasons a difference equation formulation seems to be appropriate.

On the other hand, in 1978 Bergmann [4] published experimental results on very thin film magnetization of Ni, Co and Fe deposited on a PM metallic substrate (Pb_3Bi). He interpreted his measurements to imply that the magnetization of the first few FM layers, closest to the PM, was substantially different from the bulk value. Moreover, he concluded that Ni did not develop a magnetic moment in films thinner than three monolayers. This in itself is an interesting fact, since magnetic dead layers at the surface of bulk Ni had first been reported and later disproved. Thus, Bergmann's measurements are relevant to the basic problem of the relation of magnetism and dimensionality, since in the crossover

from zero dimensions (a single magnetic Ni atom) to three-dimensional magnetic bulk Ni, the intermediate stage of a two-dimensional ultrathin Ni film deposited on a PM substrate turns out to be non-magnetic.

Bergmann's measurements constitute, to the best of my knowledge, the first experimental verification of the magnetic proximity effect (MPE); they also show unequivocally that the coherence length of $M(\vec{r})$ is of the order of magnitude of 0.1 nm.

2. METALLIC SYSTEMS

2.1. Early theories

Bergmann's experiments provided strong additional motivation for the formulation of a proper difference equation treatment of the problem. Not surprisingly shortly thereafter (1979), Cox *et al.* [5] put forward such a theoretical model to describe this experiment. Since Ni is clearly a typical example of an itinerant electron FM they suggested a treatment on the basis of a single band Hubbard model. Essentially it can be formulated as follows:

$$H = H_{FM} + H_{FM/PM} + H_{PM} \ , \tag{2.1}$$

where H_{FM}, H_{PM} and $H_{FM/PM}$ represent the ferromagnet, paramagnet and the coupling between them, respectively. They can be specified as

$$H_{FM} = \epsilon_{FM} \sum_{j,\sigma} c^{\dagger}_{j,\sigma} c_{j,\sigma} + t_{FM} \sum_{<i,j>} \sum_{\sigma} [c^{\dagger}_{i,\sigma} c_{j,\sigma} + c^{\dagger}_{j,\sigma} c_{i,\sigma}] + U \sum_{i} \sum_{\sigma} \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} \ , \tag{2.2}$$

$$H_{PM} = \epsilon_{PM} \sum_{j,\sigma} c^{\dagger}_{j,\sigma} c_{j,\sigma} + t_{PM} \sum_{<i,j>} \sum_{\sigma} [c^{\dagger}_{i,\sigma} c_{j,\sigma} + c^{\dagger}_{j,\sigma} c_{i,\sigma}] \ , \tag{2.3}$$

$$H_{FM/PM} = t_{FM/PM} \sum_{<i,j>} \sum_{\sigma} [c^{\dagger}_{i,\sigma} c_{j,\sigma} + c^{\dagger}_{j,\sigma} c_{i,\sigma}] \ . \tag{2.4}$$

Above $c^{\dagger}_{j,\sigma}$ and $c_{j,\sigma}$ are the spin σ electron creation and destruction operators at lattice site j, respectively. ϵ_{FM} and ϵ_{PM} are the FM and PM band centers, $< i, j >$ denotes only nearest neighbor summation. t_{FM}, t_{PM} and $t_{FM/PM}$ are the hopping matrix elements for the FM, PM and across the FM/PM interface, respectively, and U is the Hubbard exchange constant.

In the preceding formulation it was assumed, in order to make the problem tractable, that the interface between the FM and the PM is flat, free of defects and that the crystal structures of the FM and PM match perfectly. However, the solution of this single-band problem is non-trivial, even after the preceding simplifications are adopted. The first major difficulty that has to be overcome is the charge transfer across the interface, to which the results are extremely sensitive. In Ref. [5] this was approximated by the addition of a spin independent potential Δ added to each PM site, in order to simulate a step like dipole at the interface; next, Δ was determined self-consistently. In this way semi-quantitative conclusions on the behavior of very thin FM films deposited on a PM substrate were obtained.

The main trends that the model of Cox *et al.* [5] established are that the magnitude of the magnetic moments depend mainly on the degree of band hybridization at the interface $t_{FM/PM}/t_{PM}$ and the band filling of the FM film itself. For a monolayer, and a plausible set of hopping parameters, the model yields a loss of the Ni magnetic moment, but not so for Co or Fe. However, its major weakness is the treatment of the charge transfer across the interface.

2.2. Interface charge transfer

Not long thereafter Mata *et al.* [6] reexamined the problem carrying out a more detailed calculation of several additional physical quantities, like charge transfer and layer by layer magnetization. The system treated was a single Ni, Co or Fe atomic layer deposited on Pb_3Bi, as the typical PM. However, the major improvement introduced by Mata *et al.* was in the interfacial charge transfer treatment. This was achieved using a generalized version of the Friedel sum rule, put forward by Toulouse [7], in which the charge rearrangements are related to the scattering phase shifts. This issue is quite crucial, since small charge transfers have large physical effects. Analytically, the generalized Friedel sum rule reads as follows:

$$\Delta N = \frac{1}{\pi N} \sum_{\vec{k}} \eta(\epsilon, \vec{k}_F) = -\frac{1}{\pi N} \sum_{\vec{k}} \arg\det(\mathbf{I} - \mathbf{V}G_0) \ , \tag{2.5}$$

where ΔN is the difference between the nominal bulk electron occupation N and the FM film occupations, $\mathbf{I}$ is the unit matrix, G_0 the bulk Greens function of the PM and $\mathbf{V}$ the difference between the Hamiltonian 2.1 and the one which describes the infinite PM. The main conclusions, reached on the basis of the treatment of Ref. [6], were that both the charge

transfer and the magnetization are only significant for the FM and PM atomic layers closest to the interface.

This study was extended, using the same charge transfer treatment, to FM slabs of up to 50 monolayers thick, by Altbir *et al.* [8]. They were able to establish that for relatively weak ferromagnets, that is for $U/t_{PM} \leq 10$, the magnetization of the FM film was substantially modified, but that hardly any magnetization spread into the paramagnetic substrate was observable, not even close to the interface. These results are qualitatively independent of the filling of the single FM band.

2.3. Multiple bands

Tersoff and Falicov [9] reformulated the problem by changing the system: instead of considering a FM film on a semi-infinite PM bulk, they studied the behavior of a thin PM (3 monolayer Cu film) on a semi-infinite FM (Ni). This system has several advantages: the lattice parameters of Cu and Ni are quite similar, and both have the same crystal structure, and furthermore Cu grows well on Ni. Moreover, their treatment is more realistic, since they generalized the single band model described by Eq. 2.4, incorporating the 3d, 4s and 4p bands of Cu and Ni. A reduction to 0.33 and 0.13 μ_B of the Ni bulk magnetization (0.62 μ_B) for the nearest and second nearest layers to a (100) interface, respectively, was obtained. However, similar calculations indicated that Ni is paramagnetic (or nearly so) on the (111) face of Cu [10, 11]. Moreover, and in agreement with previous work, no appreciable penetration of the magnetization into the Cu did result.

Other authors have also explored the same ideas using closely related procedures. Mathon *et al.* [12] investigated the exchange coupling between Co and Cu, on the basis of a method put forward by Mathon [13]. Hasegawa and Herman [14] extended the calculations in two aspects: i) they studied, in a band theory formulation, the cobalt-chromium FM/AF interface, in contrast with a FM/PM one; and, ii) they included finite temperature effects.

On the experimental side of the aisle Frydman and Dynes [15], as recently as 1999, measured the magnetoresistance of granular ferromagnets suggesting that their observations could be interpreted as reflecting the existence of a magnetic proximity effect (MPE). More precisely, they measured the magnetoresistance of films of isolated Ni grains covered by different non-magnetic overlayers. They explored both the superparamagnetic to FM tran-

sition, and the impact of the different overlayers on the magnetic coupling between the Ni grains. The observation that a strong correlation of hysteresis with magnetic overlayer susceptibility does exist lead Frydman and Dynes [15] to suggest that, in high quality samples, the proximity effect may be dominant in the short distance scale. Just a few months ago Åkerman *et al.* [16] extracted, from Brillouin light scattering experiments, an upper bound for the magnetic thickness of Fe films. This upper bound turned out to be less than 1 Å for $Fe/X/ZnF_2$ and $X/Fe/ZnF_2$ systems, where X= Al and Pd, which constitutes an indication of the weakness of the MPE in metallic systems.

3. METAL-INSULATOR FM/AF INTERFACES

As a consequence of the interest that the exchange bias (EB) phenomenon [17] has attracted during the last years the interface between metallic FM's and AF insulators has become a subject of intensive research. In this context the electronic band theory models described above are replaced by a Heisenberg-like Hamiltonian descriptions of localized magnetic moments [18, 19]. Without going into specifics of the EB problem, on which we refer the reader to several recent experimental and theoretical reviews [17, 19, 20], here we will focus our attention on the difficulties related to the MPE issue that have to be overcome in the theoretical treatment of the interface. In close analogy with Eq. 2.1 the Hamiltonian that is applied in this context reads

$$\mathcal{H} = \mathcal{H}_{AF} + \mathcal{H}_{FM/AF} + \mathcal{H}_{FM} \ . \tag{3.1}$$

where $\mathcal{H}_{AF}$, $\mathcal{H}_{F/AF}$ and $\mathcal{H}_{FM}$ describe the AF substrate, interface coupling and the F slab, respectively. For the single magnetic cell, partially represented in Fig. 1, and in sharp contrast with 2.4, the localized spin Heisenberg description takes the form

$$\begin{aligned}
\mathcal{H}_{AF} = & - J_{AF} \left[S \, \hat{e}_{AF} \cdot (\vec{S}^{(\alpha)} - \vec{S}^{(\beta)}) + 2\vec{S}^{(\alpha)} \cdot \vec{S}^{(\beta)} \right] \qquad (3.2) \\
& - \frac{1}{2} K_{AF} \left[(\vec{S}^{(\alpha)} \cdot \hat{e}_{AF})^2 \ + \ (\vec{S}^{(\beta)} \cdot \hat{e}_{AF})^2 \right] - \frac{1}{2} \mu_B g \, (\vec{S}^{(\alpha)} + \vec{S}^{(\beta)}) \cdot \vec{H} \ , \\
\mathcal{H}_{FM/AF} = & - J_{F/AF} \, (\vec{S}^{(\alpha)} + \vec{S}^{(\beta)}) \cdot \vec{S}_1 \ , \qquad (3.3) \\
\mathcal{H}_{FM} = & - 2J_F \sum_{k=1}^{N-1} \vec{S}_k \cdot \vec{S}_{k+1} - \sum_{k=1}^{N} \left[\frac{K_F}{H^2} \, (\vec{S}_k \cdot \vec{H})^2 + \mu_B g \, \vec{S}_k \cdot \vec{H} \right] . \qquad (3.4)
\end{aligned}$$

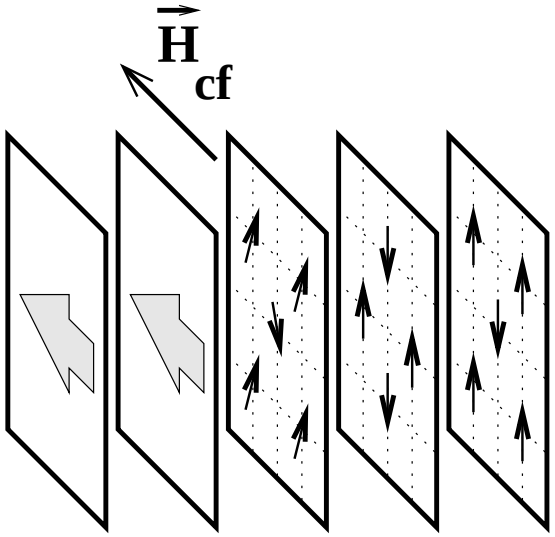

FIG. 1: Illustration of the perpendicular FM and AF magnetic interface configuration, with spin canting in the first AF layer.

Above $S = |\vec{S}|$ and N is the number of FM layers. μ_B and g denote the Bohr magneton and the Fe gyromagnetic ratio, respectively, while $\vec{H}$ is the external applied magnetic field. J_μ denotes the Heisenberg exchange parameter and K_μ the uniaxial anisotropy. In Eq. 3.2 the unit vector $\hat{e}_{AF}$ defines the AF uniaxial anisotropy direction, $\vec{S}^{(\alpha)}$ and $\vec{S}^{(\beta)}$ are canted spin vectors in the AF interface, belonging to the α- and β-AF sub-lattices.

The illustration of Fig. 1 stresses the fact, pointed out by Koon [18], that the ground state configuration corresponds to *perpendicular* orientation of the bulk FM moments relative to the AF magnetic easy axes direction. Moreover, Koon also showed that the magnetic moments in the AF interface layer exhibit canting; in fact, the minimum energy is achieved with the AF spins adopting a relatively small canting angle ($\theta < 10°$) relative to the AF bulk easy axis, with a component opposite to the cooling field $\vec{\mathrm{H}}_{\mathrm{cf}}$ direction. However this canting, which is the manifestation of the MPE, has a significant magnitude only in the AF layer closest to the interface [21–23].

The theoretical treatment outlined above actually is quite rudimentary. It assumes perfect crystallographic and magnetic order of the FM and the AF all the way to the interface, and moreover assumes perfect crystal structure matching across a flat interface. Since the MPE is only appreciable in the interface vicinity this is a rather strong assumption. Moreover, since the elastic energy related to symmetry breaking is in general so much larger than the

magnetic energies, this adds significant difficulties to a reliable treatment. Weissmann *et al.* [27], in view of the fact that both the FM and AF interface characteristics (geometry and physical parameters) are hard to determine experimentally and complicated to estimate theoretically, recently adopted two alternative interface configurations to obtain upper and lower bounds for the computed values of the exchange coupling across the interface between metallic Fe and insulating FeF_2, derived on the basis of *ab-initio* density functional theory calculations implemented for a periodic supercell. They yield values of $J_{FM/AF}$ that are quite close to each other and of the correct order of magnitude, which corresponds to ~ 1 meV. However, more realistic first principle computations of non-collinear and/or rough interfaces do not seem feasible at present.

Experimentally the MPE of metal-insulator FM/AF systems has recently received a great deal of attention. Here we limit ourselves to pointing out a few of these measurements, that are directly related to the model discussed above. Manago *et al.* [24] and Hoffmann *et al.* [25] investigated the magnetic proximity effect in NiO/Pd superlattices, with conflicting results. While the former suggested that a ferromagnetic moment of 0.59 μ_B developed per Pd atom, Hoffmann *et al.* used polarized neutron reflectometry to directly observe the Pd magnetic moment and to characterize its spatial dependence (*i.e.* the magnetization profile of the heterostructure), but without finding evidence of Pd magnetization. In addition, Hoffmann *et al.* [26] just reported neutron reflectometry results on another metal-insulator FM/AF system: $Co/LaFeO_3$, where a net magnetic moment is observed to develop in the AF layer closest to the interface. More remarkably, this AF magnetization remains constant when the magnetic field is cycled and is coupled antiferromagnetically to the FM. These experimental observations are fully compatible with the exchange bias model put forward in Refs. [21–23].

4. SPIN INJECTION

The renewed interest in the MPE is certainly related to spintronics. In fact, a century plus since the discovery of the electron, and after extensive exploitation of charge related effects, the electron spin is only now being explored for application in devices. In this context another related avenue, spin injections into semiconductors has received much attention [28, 29]. However, the physical mechanisms related to spin injection are completely different to the ones discussed above. While in the conventional MPE quantum mechanics is the governing

ingredient, spin injection is a diffusive non-equilibrium process in which electrons tunnel through a barrier at the heterojunction. The barrier is essential since otherwise, and for the reasons outlined in the preceding sections, spin injections turns out to be very weak for FM contacts with typical metallic resistivity. Present theories [28, 29] allow to conclude that spin-dependent tunneling can be employed to achieve spin injection, but that the spin injection efficiency does not exceed 2% at a temperature of 3 K [30].

Another recently explored road to spin coherence in a semiconductor is reflection off a FM. In fact, Epstein *et al.* [31] observed spontaneous spin coherence induced by FM proximity in a Mn based hybrid ferromagnet/n-GaAs heterostructure, while Ciuti *et al.* [32] analyzed theoretically the non-equilibrium spin dynamics of carriers reflected from a FM interface.

5. SUMMARY AND CONCLUSION

In summary, we have outlined the evolution of the understanding of the magnetic proximity effect (MPE), as it has developed over the last 30 years. Two fundamentally different physical systems were considered: i) a FM metal in contact with a PM metal; and, ii) a FM metal on an AF insulator. Completely distinct theoretical treatments yield, for both cases, a rather short ranged MPE. For ferromagnets on paramagnets the effect is strongest on the FM side, while for a FM on an antiferromagnet it is the AF magnetization close to the interface that is more strongly affected.

Lately, and in relation to spintronics, a different but closely related phenomenon has received much attention: the spin injection of polarized electrons into a semiconductor. This is a non-equilibrium diffusive process, of a different nature from the quantum mechanical phenomenon known as MPE. A full understanding of both, MPE and spin injection, are relevant for the exciting and challenging problem of achieving successful manipulation of electron spin in practical devices.

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