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# The indirect magnetic interaction of Fe films separated by Ag layers

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**Abstract.** We have studied Fe(110)/Ag(111)/Fe(110) and Fe(100)/Ag(100)/Fe(100) sandwiches by varying the number of Ag layers ($x$), $x = 1, \ldots, 25$, using the self-consistent real-space tight-binding method in the unrestricted Hartree–Fock approximation of the Hubbard Hamiltonian. Charge transference profiles and local magnetic moments have been calculated and the stability of the magnetic phases has been obtained. Evidence of an indirect magnetic coupling among Fe layers mediated by Ag s, p states has been found. The results are compared with previous experimental and theoretical results.

## 1. Introduction

The indirect magnetic interaction among ferromagnetic films separated by noble metal materials is now a subject of great interest [1–15]. Particularly, an oscillatory dependence of the magnetic coupling strength with the thickness of the spacer layer has been observed in a large variety of systems [4–7], creating widespread curiosity. Different theoretical models [14–19] have been proposed to explain this behaviour. Among the various combinations of metallic elements, the Fe–Ag systems have aroused considerable attention because of their immiscibility and the non-existence of intermetallic compounds.

In this paper the case of Fe(110) (Fe(100)) films separated by a variable number of monolayers of Ag(111) (Ag(100)) will be addressed. This problem has been studied experimentally and theoretically by several techniques and authors [8–14]. In particular, it has been established, using transmission Mössbauer spectroscopy [10] that 3D behaviour is present when the number of Ag spacer monolayers (ML) is less than 15. Also, inverse photoemission experiments [11, 12] have shown the appearance of a periodicity in the measured inverse photoemission peaks, plotted as a function of increasing Ag thickness, with a period of about 5 ML. Finally, an oscillatory exchange magnetic coupling in Fe/Ag(100) systems has been measured in ferromagnetic resonance (FMR) and SMOKE experiments [13], and has been predicted by RKKY calculations [14].

This work was done with the purpose of clarifying the role played by the Ag spacer layers, using a mean-field model of itinerant electrons. Within this simple model, we calculated local magnetic moments and charge transfer between Fe and Ag layers and we have established the preferred magnetic order for different Ag thicknesses. Analysing the obtained results, evidence has been found of an indirect magnetic coupling among the Fe films across the Ag spacer layers, which is mediated by the s, p Ag orbitals, reaching up to

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15 ML. An oscillatory behaviour of the value of the density of states per atom at the Fermi level and of the interlayer magnetic coupling, both of them with a periodicity of 5 Ag ML, has also been observed.

The outline of the present paper is as follows: section 2 is devoted to a short description of the self-consistent real-space tight-binding method in the unrestricted Hartree–Fock approximation to the Hubbard Hamiltonian [20] employed in this study. In section 3, the obtained results are presented and compared with those previously reported [8–14]. Finally, in section 4 we display our conclusions.

## 2. The model and parameters

The magnetic coupling observed in previous works [7, 15] depends mainly on the thickness of the noble metal spacer layers. Considering this, and to better the understanding of how the Ag thickness affects the magnetic coupling, we adopt $(Fe)_1/(Ag)_x/(Fe)_1$ sandwiches ($x = 1, \ldots, 25$). We neglect the crystalline anisotropy in order to simplify our calculations, although it may play an important role. In order to do this we consider that the Fe monolayers have the same crystalline structure as the Ag spacer layers.

To study the system theoretically the following Hubbard-type Hamiltonian is employed

$$H = \sum_{i,m,\sigma} E_{i,m} N_{i,m,\sigma} + \sum_{i \neq j,m,m',\sigma} T_{i,j}^{m,m'} a_{i,m,\sigma}^{\dagger} a_{j,m',\sigma} + H_{ee} \tag{1}$$

where $a_{i,m,\sigma}^{\dagger}$, $a_{i,m,\sigma}$ and $N_{i,m} = a_{i,m,\sigma}^{\dagger} a_{i,m,\sigma}$ refer to the creation, annihilation and number operator of an electron at atomic site $i$ in the orbital $m$ with spin $\sigma$ ($\sigma = 1$: majority, $\sigma = -1$: minority). $E_{i,m}$ stands for the $m$th energy level at site $i$ and $T_{i,j}^{m,m'}$ for the hopping integral between sites $i$ and $j$.

The electron–electron interaction Hamiltonian can be written following [21] as:

$$H_{ee} = \frac{1}{2} \sum_{i,m} U_{i,m,m} N_{i,m} + \frac{1}{2} \sum_{i,m,m'} U_{i,m,m'} N_{i,m} N_{i,m'} - \sum_{i,m,m'} J_{i,m,m'} \mathbf{S}_{i,m} \cdot \mathbf{S}_{i,m'} \tag{2}$$

where the spin operator $\mathbf{S}_{i,m} = \frac{1}{2} \sum_{\alpha,\beta} a_{i,m,\alpha}^{\dagger} \sigma_{\alpha,\beta} a_{i,m,\beta}$ ($\sigma_{\alpha,\beta}$ are the Pauli matrices), and $N_{i,m} = \sum_{\sigma} N_{i,m,\sigma}$.

Treating (1) in the unrestricted Hartree–Fock approximation and using a self-consistent mean-field magnetization $M_{i,m} = \langle \mathbf{S}_{i,m} \rangle$ the last term of (2) becomes

$$\sum_{i,m,m'} J_{i,m,m'} (\mathbf{S}_{i,m} \cdot M_{i,m'} + M_{i,m} \cdot \mathbf{S}_{i,m'} - M_{i,m} \cdot M_{i,m'}). \tag{3}$$

Within the present model s, p and d orbitals are considered, and in the hopping integrals up to second-nearest-neighbour interactions are included. $E_{i,m}$ and $T_{i,j}^{m,m'}$, for Fe–Fe and Ag–Ag interactions, are calculated following [22]. As for $T_{i,j}^{m,m'}$ between Fe and Ag, we employ the geometrical averages of the values of Fe–Fe and Ag–Ag pairs [23, 24].

The hopping integrals are dependent on the distance between atoms. In order to neglect anisotropy it is necessary to modify the Fe lattice constant, this change must be considered when Fe–Fe interactions are calculated. The bcc Fe(100) surface matches the fcc Ag(100) surface to within 1%, so in this case we decided to employ the Fe bulk lattice parameter. In other cases, we needed to expand the Fe lattice constant so that the bcc Fe(110) layers coincided with the fcc Ag(111) structure.

In $H_{ee}$ only the effective intra-atomic Coulomb repulsions, $U_{i,m,m'}$, are used. These are not the atomic ones but are renormalized by correlation effects, as in the work by

Sarma [25]. Their values are summarized in table 1. The exchange integrals, $J_{i,m,m'}$, are considered to be non-zero only for Fe d orbitals. The latter was fitted in order to give 2.3 $\mu_b$ magnetization for a Fe bulk; its resulting value $J_{d,d} = 0.97$ eV.

**Table 1.** The effective intra-atomic Coulomb repulsion $U_{i,m,m'}$. where $m$, $m'$ stand for s or p and $U_{m,d} = U_{d,m}$.

| | Fe | Ag |
|---|---|---|
| $U_{m,m'}$ | 0.754 eV | 0.745 eV |
| $U_{m,d}$ | 0.971 eV | 0.958 eV |
| $U_{d,d}$ | 2.33 eV | 2.15 eV |

The spin-polarized local density of states (LDOS)

$$\rho_{i,\sigma} = \sum_m \rho_{i,m,\sigma} = -\frac{1}{\pi} \sum_m \mathrm{Im}\{G_{im\sigma,im\sigma}\} \tag{4}$$

is determined by calculating the local Green function $G_{im\sigma,im\sigma}$ by means of the self-consistent real-space recurrence method [26]. The number of $k$ levels of the continuous fraction expansion of $G_{im\sigma,im\sigma}$ is chosen to be large enough that the results become independent of $k$. Empirically, it was found that $k = 12$ fulfils this requirement. This number of levels comprises a compromise between computing time, which grows extensively when the numbers of levels increase, and the convergence of the resulting self-consistent value of the magnetic moment.

The electronic occupation $N_{i,m}$ and the local magnetic moment $M_{i,m}$ at site $i$ and orbital $m$, are given by

$$M_{i,m} = \langle N_{i,m,\uparrow} \rangle - \langle N_{i,m,\downarrow} \rangle \tag{5}$$

and

$$N_{i,m} = \langle N_{i,m,\uparrow} \rangle + \langle N_{i,m,\downarrow} \rangle \tag{6}$$

which are determined self-consistently by requiring $\langle N_{i,m,\sigma} \rangle = \int_{-\infty}^{E_F} \rho_{i,m,\sigma}(E)\,\mathrm{d}E$ where the Fermi level ($E_F$) is determined by demanding global charge neutrality. The self-consistent procedure is started by taking the bulk values as initial values for $N_{i,m}$ and $M_{i,m}$ and is stopped when the absolute value of the difference between the output and input values is less than $10^{-4}$. Therefore, when the convergence is reached, the charge transference, $\Delta N_i = \sum_m N_{i,m}(\mathrm{final}) - N_{i,m}(\mathrm{initial})$, and magnetic moments, $M_i = \sum_m M_{i,m}$, are obtained layer by layer.

## 3. Results and discussion

Within the present model we have found that the magnetization and electronic occupations of Fe monolayers separated by a variable number of Ag monolayers depends on Ag thickness. Figure 1 shows the local magnetic moment of Fe as a function of the Ag thickness for both cases and it is clear that at approximately 15 ML a limiting value is reached. This limiting value, 2.85 $\mu_b$ (2.95 $\mu_b$), is equal to that obtained for a Fe(110) (Fe(100)) layer on an infinite Ag(111) (Ag(100)) substrate.

Due to the assumption of global charge neutrality there is an increasing charge transference from Fe to Ag that reaches a limiting value of 0.3 electrons in the case of Fe(110), for the same number of Ag monolayers for which the limiting magnetization value

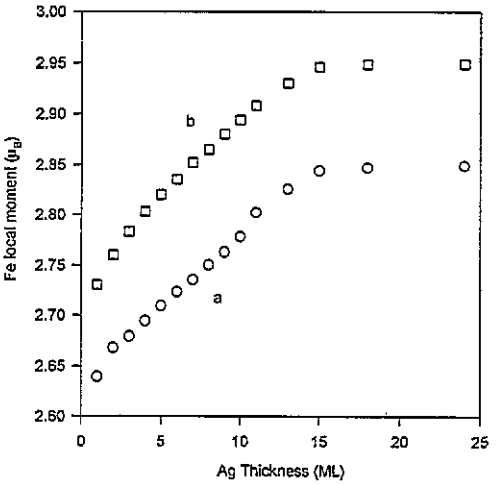


**Figure 1.** The calculated Fe local magnetic moment versus Ag thickness for (*a*) Fe(110)/Ag(111)/Fe(110) (○) and (*b*) Fe(100)/Ag(100)/Fe(100) (□).

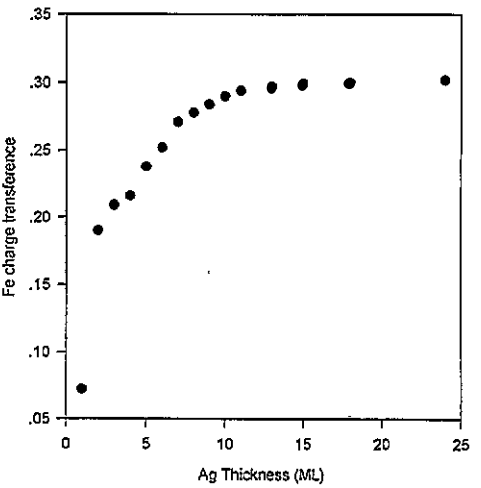


**Figure 2.** The calculated Fe charge transference versus Ag thickness for Fe(110)/Ag(111)/Fe(110) trilayers.

is obtained. Figure 2 shows the total charge transference for Fe(110) as a function of Ag thickness. Most of the charge transference goes from Fe d minority orbitals to Ag s, p orbitals.

We have considered both, ferromagnetic and antiferromagnetic couplings. The stabilities of each magnetic phase, for different Ag thicknesses, were obtained by calculating the total energy in the Hartree–Fock approximation, using the expression given by Boubard *et al* [27]. This total energy is the sum over the one-electron states corrected by the Coulomb and exchange energy counted twice in this sum. This is sufficient to differentiate, at a fixed lattice position, between ferromagnetic and antiferromagnetic coupling.

Having computed the ferromagnetic energy ($E(FM)$) and the antiferromagnetic energy ($E(AFM)$) separately, we subtract them and plot this as function of the Ag thickness. Doing this we have found that, $\Delta E = E(FM) - E(AFM)$ oscillates around zero. Therefore the system prefers ferromagnetic and antiferromagnetic coupling alternatively up to 15 ML of Ag thickness; from there on the numerical accuracy of our calculations is not sufficient to differentiate between the magnetic couplings. This is shown in figure 3, for the Fe(100)/Ag(100)/Fe(100) system, where it is possible to see that the spacing corresponding to each coupling is approximately 2.5 ML and that the period of oscillations is 5 ML. This is similar to the exchange coupling periodicity reported by Celinski *et al* [13] (5–6 ML) and the long wavelength period calculated by Bruno and Champert [14] (5.6 ML) using an extended RKKY theory.

One may well ask why the present model gives a period different from that of RKKY. It is known that the period of the oscillations in the exchange coupling is determined by topological properties of the Fermi surface in the spacer layer [12, 14, 16]. We think that the discrepancy between the present results and those previously reported [14] can be attributed to a possible inaccuracy of our Fermi surface description.

Himpsel and Ortega [11, 12] have reported that, when they plotted the peaks of the inverse photoemission spectrum near $E_F$, for Fe/Ag(100) superlattices as a function of Ag thickness, they obtained a periodicity of five layers. They associate this periodicity with

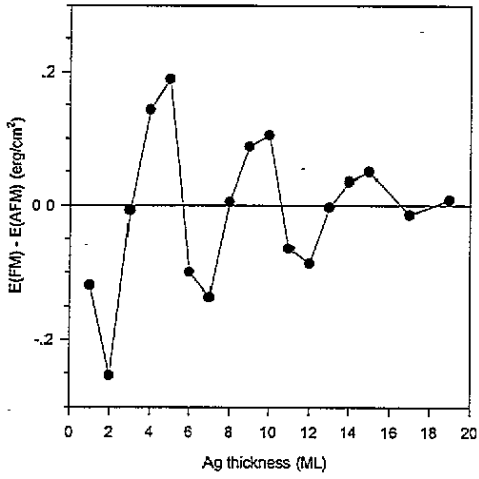


**Figure 3.** $E(FM) - E(AFM)$ as a function of Ag thickness for Fe(100)/Ag(100)/Fe(100). Positive values indicate antiferromagnetic coupling and negative values ferromagnetic coupling.

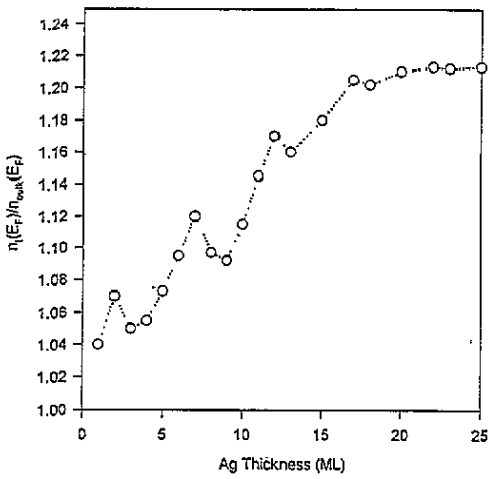


**Figure 4.** $n_t(E_F)/n_{bulk}(E_F)$ versus Ag thickness for Fe(100)/Ag(100)/Fe(100) sandwiches.

the existence of quantum-well states, having an s–p character, and connect them with the oscillatory magnetic coupling of this system. As the measured intensity in a photoemission experiment is proportional to the density of states near the Fermi level, we calculated the total density of states at $E_F$ and plotted it as function of a number of Ag monolayers in order to compare with their results. Again an oscillating behaviour is obtained with a periodicity of 5 ML; it is shown in figure 4, for the case Fe(100)/Ag(100)/Fe(100), where the value of the total density of states at $E_F$ as a function of Ag thickness normalized with the value of the density of states at $E_F$ of Fe bulk is plotted.

When analysing the origin of the oscillations appearing in the density of states, the s, p occupation of the silver layers, $N_{s,p}$, was plotted for different Ag widths and it was found that $N_{s,p}$, for the cases which correspond to maxima of the density of states at $E_F$, resemble stationary quantum-well states. In addition the silver s, p states are found to be spin polarized despite their non-magnetic s, p character, thus providing an indirect magnetic interaction. The origin of the spin polarization of these states lies on the spin-dependent boundary condition at the interface with the Fe layers. In this way our model couples the magnetic layers by polarizing the non-magnetic layers, this is precisely the situation addressed by the RKKY theory. It is important to note that an hybridization between Fe and Ag states occurs at the interfaces via the hopping integrals. This hybridization made the spin polarization of the non-magnetic silver states and the charge transference from Fe to Ag possible.

It can also be observed in the Fe local density of states that, at the Fermi level, the Fe majority band is nearly zero and the Fe minority band has pronounced peaks corresponding to the d orbitals, see figure 5. The Fermi energy moves around these peaks when increasing the number of Ag monolayers. A direct consequence of this is the growing charge transference from the minority Fe d band to the Ag s, p bands, which is responsible for the increasing local magnetic moment.

Finally, it is interesting to compare results obtained by Walker *et al* [8, 10] with our

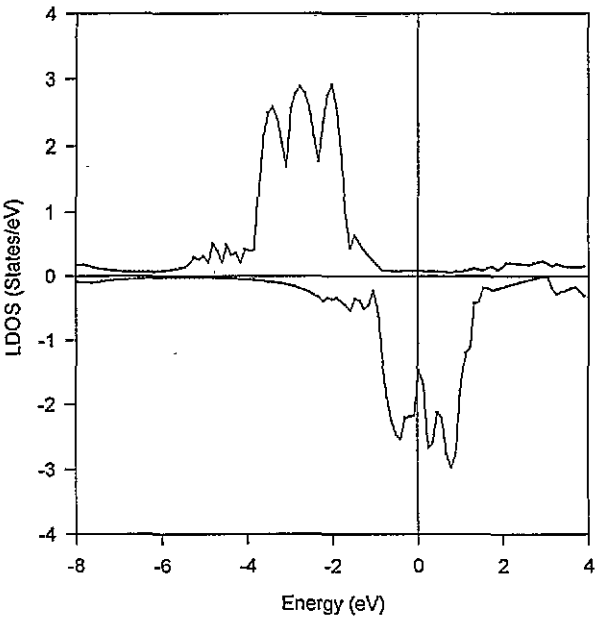


**Figure 5.** Fe local density of states (LDOS) for Fe(100)/10Ag(100)/Fe(100). Shown are the LDOS for the majority (positive values) and minority (negative values) spin directions.

results. In references [8] and [10] a transition from 3D to 2D magnetic behaviour is observed when going beyond 15 Ag ML, as a consequence of the varying strength of the indirect magnetic coupling between Fe layers. In this work it was found that, when the silver thickness is around 15 ML, the oscillations shown in figures 3 and 4 disappear and the charge transference and Fe local magnetic moment reach a constant value. The latter is particularly like the result obtained for a Fe monolayer on a Ag subtract. There is also evidence that Fe layers stop 'seeing' themselves across the noble metal spacer layer at around 15 ML of Ag.

## 4. Conclusions

Summing up, within the unrestricted Hartree–Fock approximation and the framework of itinerant ferromagnetism the existence of an indirect magnetic coupling between Fe layers is verified. The indirect exchange coupling between Fe layers occurs due to the hybridization of Fe and Ag states at the interfaces. This hybridization occurs via the hopping matrix elements $T_{i,j}^{m,m'}$ which connect states in the Fe and Ag planes and polarizes the silver states, leading to an indirect coupling. One consequence of this spin polarization of the silver states is the existence of charge transference among the Fe minority orbitals and the Ag s, p orbitals.

The obtained range of the interaction among Fe layers through Ag is similar to the range observed experimentally by Walker [8, 10]. The diappearance of the indirect magnetic interaction is evidenced by the missing oscillations in figures 3 and 4 and by the reaching of limiting values of charge transference and magnetic local moment. Our results concerning the periodicity are directly comparable with the previous reported results [12–14]. The difference between the period obtained in this work and that predicted from RKKY calculations [14] is due to a possible inaccuracy in our Fermi surface description.

In conclusion, the tight-binding scheme, within its flexibility, allows us to understand qualitatively the origin of the indirect magnetic interaction between Fe layers within a reasonable computing time. It is clear from our results that there exists an indirect magnetic coupling between Fe layers which is mediated by those Ag orbitals which are mostly of s, p character. In order to improve the present results, we are now considering the use of finer meshes in the energy integration and more steps in the continuous fraction, although it will take much more computing time. However, a profound understanding of the properties of these systems requires further, more sophisticated studies.

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