Interlayer Coupling in Magnetic/Pd Multilayers

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Abstract
The Anderson model of local-state conduction electron mixing is applied to the problem of interlayer magnetic coupling in metallic multilayered structures with palladium (Pd) spacer layers. An oscillation period of 5 spacer monolayers and the tendency towards ferromagnetic bias of the interlayer magnetic coupling that we obtain are consistent with the experimental data.

The discovery of oscillating interlayer magnetic couplings between ferromagnetic layers separated by a nonmagnetic metallic spacer [1] and of the related giant magnetoresistance effect [2], has stimulated a lot of experimental and theoretical activity. It has been shown that the periods of the coupling are related to the topology of the Fermi surface of the spacer layers. This interpretation has been confirmed by model and first-principles calculations, and is also supported by experiments [3, 4]. There are, however, other aspects of the coupling, e. g., the bias (ferro- or antiferro-magnetic) of the interlayer magnetic coupling, which have not been fully explained.

For Fe(001) layers separated by Pd(001) spacers of thickness between 4 and 12 ML the interlayer magnetic coupling is observed to have a strong ferromagnetic bias as seen in the experiments [5]. Above a 13 ML thickness of the Pd spacer the coupling begins to be antiferromagnetic. Metallic Pd is believed to be near the threshold of becoming ferromagnetic. The non-relativistic calculations of Moruzzi and Marcus [6] and of Chen et al. [7] predicted the onset of ferromagnetism in fcc palladium with a 5% expanded lattice. In a recent publication the ferromagnetic bias of the coupling in magnetic multilayer structures with a Pd spacer is explained in terms of the Pd as an almost ferromagnetic media [8]. Alternatively, in this paper, we interpret this ferromagnetic bias to be a consequence of a competition between RKKY-like and superexchange couplings, with RKKY coupling being dominant.

The RKKY-like coupling comes from intermediate states which correspond to spin excitations of the Fermi sea. States corresponding to electron-hole pair production in the Fermi sea, with an attendant spin-flip, contribute to the RKKY coupling as [8]:

$$j_{RKKY}(q) = \sum_{n_1,n_2,k} \left| V_{n_1k} \right|^2 \left| V_{n_2k'} \right|^2 \frac{\theta(\varepsilon_F - \varepsilon_{n_1k})\theta(\varepsilon_{n_2k'} - \varepsilon_F)}{(\varepsilon_{n_2k'} - \varepsilon_{n_1k})^2} + c.c.,$$

where $\theta$ is a step function, $\varepsilon_F$ is the Fermi energy, $k' = k + q + G$, $G$ is a vector of the reciprocal lattice, $\varepsilon_\pm$ is the energy of the local impurity state, and $V_{nk}$ represents the strength of the $s - d$ mixing interaction [8].
The superexchange coupling arises from charge excitations in which electrons from local states are promoted above the Fermi sea (one from each layer) providing a second contribution to the coupling in addition to the RKKY coupling [8]:

\[
\hat{j}_S(q) = - \sum_{n_1, n_2, k} \left[ \frac{|V_{n_1 k}|^2 |V_{n_2 k'}|^2}{(\varepsilon_{n_2 k'} - \varepsilon_+)^2} \frac{\theta(\varepsilon_{n_1 k} - \varepsilon_F)\theta(\varepsilon_{n_2 k'} - \varepsilon_F)}{\varepsilon_{n_1 k} - \varepsilon_+} + \text{c.c.} \right].
\]

(2)

The real space coupling between two sheets of spins can be obtained by Fourier transforming Eqs. (1) and (2) [8], with the coupling in the real space given by,

\[
J_l(z) = \frac{a}{2\pi} \int_0^\infty dq_z \hat{j}(q_z) \cos(q_z z),
\]

(3)

where \(a\) is a lattice constant, and \(z\) is in the direction perpendicular to the magnetic layers. The sign is chosen so that positive \(J_l(z)\) signifies ferromagnetic coupling.

Using the Slater-Koster parameters [11], one can easily diagonalize small matrices (9x9 for a typical transition metal) to obtain the energy bands and density of states (DOS) for fcc Pd. The electron wave function \(|n, k\rangle\) is a Bloch state belonging to band \(n\) and wave vector \(k\), and is expressed as linear combinations of localized orbitals:

\[
|n, k\rangle = \frac{1}{\sqrt{N}} \sum_\nu \sum_i a_{ni}(k) u_i(r - R_\nu),
\]

(4)

where \(N\) is the number of cells in the material considered, \(R_\nu\) is a lattice vector, \(u_i(r - R_\nu)\) is the \(i\)th orbital basis function, and \(a_{ni}(k)\) is a (real) normalized eigenvector component determined by diagonalization of the single-particle Hamiltonian. We use a plausible approximation \(V_{n_1 k}V_{n_2 k'}^* = V^2 M_{n_1 k, n_2 k'}(q)\) [3], where the matrix element is defined as \(M_{n_1 k, n_2 k'} = \langle n_1 k | e^{i\mathbf{q}\cdot\mathbf{r}} | n_2 k'\rangle\). The explicit expression for the matrix element is

\[
M_{n_1 k, n_2 k'} = \sum_\nu \sum_{i,j} e^{ikR_\nu} a_{ni}(k) a_{nj}(k') \int d\mathbf{r} u_i(r) e^{i\mathbf{q}\cdot\mathbf{r}} u_j(r - \mathbf{R}_\nu).
\]

(5)

The essential conditions for the simplification of this matrix was already discussed by Callaway et al. [1], and \(u_i(r)\) are approximated as Clementi wave functions for the d states [1], and \(a_{ni}(k)\) can be related to the Slater-Koster parameters in Ref. [10].

We consider one local level below \(\varepsilon_F\) for simplicity and set \(\varepsilon_+ = \varepsilon_F - E_h\), where \(E_h\) is the energy required to promote an electron from an occupied local magnetic impurity level to the Fermi level. Based on the band structure of bulk paramagnetic Pd, we have calculated the couplings \(j_{RKKY}(q_z)\) and \(j_S(q_z)\) with \(E_h = 0.08\) Ry, as shown in Fig. 1.

The couplings in real space are plotted in Fig. 2. The dashed line, dotted line and solid line are for RKKY-like, superexchange, and RKKY + Superexchange couplings, respectively. We see that the superexchange interaction gives a small contribution to the
coupling, and the total coupling has a strong ferromagnetic bias. This tendency for a ferromagnetic bias resembles the experimental observation in Fe/Pd(001) trilayered structures [9]. The 5 ML oscillation period in the calculated interlayer magnetic coupling $J(z)$, as shown in Fig. 2, corresponds to the peak at $q_z \approx 0.4 \frac{2\pi}{a}$ in $j_{RKKY}(q_z)$. It agrees with the experimental period of 4 – 5 ML in Fe/Pd/Fe(001) for trilayered structures [9].

We explain the result of ferromagnetic bias for multilayers with Pd spacers as being due to the structure of the Pd DOS and the location of the Fermi level, as shown in Fig. 3. In particular, the fact that the Fermi level falls above a peak in the DOS followed by a relatively smooth and structureless DOS, results in a relatively small superexchange contribution above $\sim 4$ ML, and leads to a ferromagnetic bias driven by the large RKKY coupling.

To confirm our explanation of the cause of the ferromagnetic bias we use a free-electron gas model which enables us to obtain analytic results for the couplings. In a previous study [12], we showed that in the free-electron gas approximation, RKKY + Superexchange coupling resembles pure RKKY coupling, but without any magnetic bias. To illustrate the effect of a peak in the density of states on top of a free electron-like background, we use a “toy model” calculation by adding a Lorentzian shaped peak to the DOS of the free electron gas,

$$D(E) = \sqrt{E + \frac{\sqrt{E_F}}{(E/\varepsilon_F - p)^2 + h^2}},$$

(6)

where the position of the peak is at $E_p = p \varepsilon_F$, and $h$ adjusts height of the peak (small $h$ corresponds to a large peak). For example, by fixing the position ($p = 0.9$, below the Fermi level) and increasing the height of the peak (e.g., $h = 0.3$), ferromagnetic bias occurs in the coupling, as shown in Fig. 4. We noted that in multilayer structures with a Cr spacer, RKKY + superexchange coupling gives an antiferromagnetic bias due to the structure of the DOS, with a peak above, but near to the Fermi level $\varepsilon_F$ [12]. This is also confirmed in our “toy model” calculations.

The coupling observed in the experiments contains a bilinear exchange interaction $J_1$ and a biquadratic exchange interaction $J_2$. In conventional notation, $J_{exp} = J_1 - 2J_2$ [3]. Here, RKKY-like and superexchange couplings are contained in the bilinear coupling, $J_1$.
Figure 5: Interlayer magnetic coupling as a function of Pd spacer thickness. The solid line and filled circles are our calculations and the experimental data observed at $T = 77\, \text{K}$, respectively. The theoretical (experimental) results are referred to the left (right) scales.

and a positive $J_2$ favors a perpendicular magnetic coupling. The ratio of $J_2/J_1$ observed in the multilayer structures with Cr spacers is about $0.3 - 0.5$. Magnetic multilayer structures with Pd spacers also have a relatively large biquadratic exchange interaction $J_2$. With a proper choice of positive $J_2$ ($J_2/J_1 \approx 0.5$ at $13\, \text{ML}$), the bias can switch to antiferromagnetic for spacer thickness greater than $13\, \text{ML}$, as is observed experimentally. One can see that our calculated interlayer magnetic coupling with Pd spacers can be used to explain the experimental data as shown in Fig. 5.

In summary, our model calculation has been able to reproduce the two salient features of the interlayer magnetic coupling in Fe/Pd(001) multilayer structures: large but rapidly decreasing ferromagnetic bias, and a $5\, \text{ML}$ oscillation period. The ferromagnetic bias arises from the competition between the RKKY-like and superexchange couplings due to the special features of the palladium DOS: relatively large peak below, but near to the Fermi level, and a small DOS above the Fermi level.

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