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Effects of impurities on the exchange coupling in magnetic metallic multilayers

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Abstract
In this work the variation of the exchange coupling between magnetic layers across a non-magnetic spacer layer, due to impurities of the magnetic material in the spacer, is calculated. The impurities are treated in the dilute limit and beyond it. It is shown that the change per impurity is comparable to the unperturbed coupling per atom, and that it oscillates as a function of spacer layer thickness with the same period as that coupling, though with a different phase. The most favourable magnetic moment orientation of the impurity is found for some spacer layer thicknesses and impurity positions. It is also shown that the short-period oscillations of the coupling may be much more affected by the presence of impurities than the long-period ones.

1. Introduction

Oscillations in the exchange coupling between layers of a metallic ferromagnet across a non-magnetic metal were first observed about one decade ago by Parkin *et al* [1]. This phenomenon has attracted a great deal of attention and has become the subject of intensive research activity since then. Significant progress has been made towards the understanding of the basic mechanism responsible for the effect (for a comprehensive review see reference [2]). However, the poor agreement between experimental and theoretical results for the exchange coupling in metallic multilayers, in particular with respect to the oscillation amplitudes, has been a matter of concern for both theoreticians and experimentalists. The source of disagreement is usually attributed to deviations from the idealized situation often assumed in theoretical works. For example, the existence of sharp interfaces and of perfect crystallinity in the layers are assumptions underlying most of the theoretical studies of the coupling. Real systems always exhibit some degree of imperfections, and this has been regarded as the main source of the discrepancies between measurements and calculations. In fact, the role

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of interface quality in the oscillatory coupling has been clearly demonstrated by Unguris *et al* [4].

The precise state of the interfaces in real samples cannot be easily characterized experimentally. Therefore, it is important to investigate how different types of deviation from perfectly flat interfaces affect the coupling. Edwards *et al* [21] have considered interface structures consisting of plateaus periodically arranged along one of the directions parallel to the interface. An alternative approach adopted by several authors [3, 6, 18] is based on the calculation of configuration averages of the coupling over different interface profiles. This is achieved by regarding the interface region as consisting of an alloy of the magnetic and non-magnetic materials, with suitable composition, and by calculating the coupling within the coherent potential approximation. It is worth pointing out that the introduction of the configuration average recovers to some extent the translation symmetry of the system parallel to the atomic planes, reducing the effects of incoherent scattering at the interface. As regards deviations from crystallinity within the spacer layer, very little has been done from the theoretical point of view.

Here we investigate how the exchange coupling is affected by magnetic impurities localized in the spacer, both in the interface region and well within the spacer. We first deal with single magnetic impurities (diluted regime), and next consider the case of plateaus of magnetic atoms at the interface between one of the magnetic layers and the spacer. In the case of impurities inside the spacer, the orientation of the impurity magnetization relative to those of the ferromagnetic layers is determined so as to minimize the total energy of the system. Calculations are performed for two spacer band fillings, corresponding to long- and short-period exchange-coupling oscillations. We find that the effects of the presence of magnetic atoms in the spacer are more important in the latter case, and increase with the size of the clusters.

2. Theory

Our system consists of two parallel atomic layers of a ferromagnetic metal embedded in a non-magnetic material. We label the two ferromagnetic layers 0 and $N+1$, so the spacer thickness is equal to N atomic planes. For simplicity, we shall use a single-orbital tight-binding model with nearest-neighbour hoppings only, on a simple cubic lattice. We assume that the exchange interaction within the magnetic material is described by a local exchange potential V_{ex}.

We first deal with the case of a single impurity atom of the magnetic layer material placed in the spacer at an arbitrary site in plane l, where $0 < l < N + 1$. To calculate the coupling between the ferromagnetic layers, we evaluate the energy necessary to rotate the magnetization $\hat{m}_{\rm B}$ of layer $N + 1$ relative to that of layer 0 ($\hat{m}_{\rm A}$). We assume that the impurity magnetic moment has the same value as the magnetization per atom of the magnetic layers. For various spacer thicknesses and impurity positions we consider, in each case, four possible magnetic arrangements of the system, namely, the magnetic impurity moment $\hat{m}_i$ may be either parallel or anti-parallel to $\hat{m}_{\rm A}$, as represented in figure 1. Total-energy differences between these states have been calculated and the most stable magnetic configuration for the impurity is determined for each arrangement of the ferromagnetic layers. The interlayer coupling was then obtained by calculating the work necessary to rotate the magnetization of the ferromagnetic layers from parallel to anti-parallel.

First we consider the change in the coupling caused by a single impurity in the spacer, and then the effects due to the presence of magnetic plateaus at the interface.

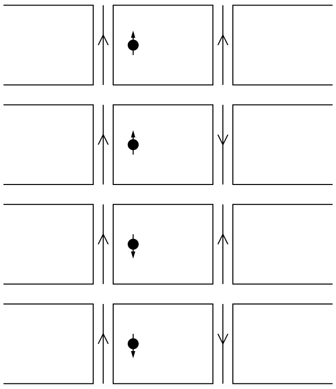

Figure 1. A schematic view of the four possible arrangements of the system with one impurity. The lines with arrows represent the magnetic planes and the filled circles with arrows represent the impurity; the arrows denote the direction of the local magnetization. The closed polygon regions represent the non-magnetic material.

2.1. Impurity at the interface

We start by considering an extra atom of the magnetic material in plane 1 of the spacer, next to ferromagnetic plane 0. In this case, due to the proximity between the impurity atom and plane 0, it is reasonable to assume that the impurity magnetic moment remains parallel to $\hat{m}_{\rm A}$ no matter what the orientation of $\hat{m}_{\rm B}$ is relative to $\hat{m}_{\rm A}$. The work necessary to rotate $\hat{m}_{\rm B}$ by an angle θ with respect to $\hat{m}_{\rm A}$ is given by

$$\Delta\Omega(\theta) = \Omega(\theta) - \Omega(0) \tag{1}$$

where $\Omega(\theta)$ is the thermodynamic potential of the system. We can write

$$\Omega(\theta) = \Omega^{(0)}(\theta) + \delta\Omega(\theta) \tag{2}$$

where the superscript (0) refers to the system without the impurity atom, and $\delta\Omega(\theta)$ corresponds to the change in the thermodynamic potential due to the introduction of the impurity atom. The interlayer coupling is defined as $J = \Delta\Omega(\pi)$, and so we have that

$$J = J^{(0)} + \delta J. \tag{3}$$

$J^{(0)}$ can be obtained using several very general methods [13, 15, 16], so the evaluation of the effects of impurity on the coupling rests on the calculation of δJ. This can be done as described below.

We assume that the impurity perturbing potential $\hat{V}$ is restricted to the impurity site and is given by

$$\hat{V} = v|1, \vec{0}\rangle\langle 1, \vec{0}| \tag{4}$$

where $|1, \vec{0}\rangle$ is the atomic state localized on site $\vec{R} = \vec{0}$ of plane $l = 1$, and $v = V_0 I - V_{ex}\sigma_z$. Here, V_0 is the non-magnetic potential difference between the spacer and the magnetic layer material, V_{ex} is the strength of the local exchange potential at the impurity site, I is the identity matrix in spin space, and σ_z is the usual Pauli matrix. It follows that

$$\delta\Omega(\theta) = -\frac{1}{\beta}\int_{-\infty}^{+\infty} \mathrm{d}\omega\ \ln(1 + \mathrm{e}^{\beta(\mu-\omega)})\,\delta\mathcal{D}(\omega, \theta) \tag{5}$$

where $\delta\mathcal{D}(\omega, \theta)$ is the corresponding change in the density of states of the system due to the introduction of the impurity, $\beta = 1/k_B T$, and μ is the chemical potential. The change in the density of states $\delta\mathcal{D}$ is given by

$$\delta\mathcal{D}(\omega, \theta) = -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \sum_j \delta G_{jj}(\omega, \theta) \tag{6}$$

where 'Tr' stands for the trace over spin and atomic orbital indices, the sum is over all atomic sites $j = (l, \vec{R})$, and

$$\delta G_{jj}(\omega, \theta) = G_{jj}(\omega, \theta) - G^{(0)}_{jj}(\omega, \theta) \tag{7}$$

is the change in the site-diagonal matrix elements of the one-electron Green function due to the impurity. Using Dyson's equation in operator notation, we find

$$\delta\hat{G}(\omega, \theta) = \hat{G}^{(0)}(\omega, \theta)\hat{T}(\omega, \theta)\hat{G}^{(0)}(\omega, \theta) \tag{8}$$

where $\hat{T} = \hat{V}(1 - \hat{G}^{(0)}(\theta)\hat{V})^{-1}$ is scattering-matrix operator. Due to the local character of the impurity potential, it is straightforward to show that

$$\delta\Omega(\theta) = \frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{+\infty} \mathrm{d}\omega\, f(\omega) \ln\left[1 - G^{(0)}_{1,\vec{0};1,\vec{0}}(\omega, \theta) v\right] \tag{9}$$

where $f(\omega)$ is the Fermi–Dirac distribution function and

$$G^{(0)}_{1,\vec{0};1,\vec{0}}(\omega, \theta) = \langle 1, \vec{0}|G^{(0)}(\omega, \theta)|1, \vec{0}\rangle.$$

After some algebra, we obtain the impurity contribution to the coupling as given by

$$\delta J = \frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{+\infty} \mathrm{d}\omega\, f(\omega) \ln\left[\Lambda(\omega, \pi)\Lambda^{-1}(\omega, 0)\right] \tag{10}$$

where $\Lambda(\omega, \theta) = 1 - G^{(0)}_{1,\vec{0};1,\vec{0}}(\omega, \theta)v$.

In view of the in-plane translational invariance of the unperturbed system, it is convenient to express $G^{(0)}_{1,\vec{0};1,\vec{0}}(\omega, \theta)$ in terms of the plane-diagonal matrix elements of the unperturbed Green function in the mixed representation $G^{(0)}_{11}(\vec{k}_\parallel, \omega, \theta)$ as

$$G^{(0)}_{1,\vec{0};1,\vec{0}}(\omega, \theta) = \sum_{\vec{k}_\parallel} G^{(0)}_{11}(\vec{k}_\parallel, \omega, \theta) \tag{11}$$

where $\vec{k}_\parallel$ is a wave vector parallel to the atomic planes belonging to the two-dimensional Brillouin zone.

2.2. *Impurity anywhere in the spacer*

As regards the case in which the impurity occupies an arbitrary position in the spacer layer, the theory described above has to be changed so as to allow the impurity magnetic moment to point either up or down relative to $\hat{m}_0$, depending on whether the configuration of the magnetic layers is ferromagnetic ($\theta = 0$) or antiferromagnetic ($\theta = \pi$). This can be achieved by introducing the perturbing potential

$$\hat{V}_\xi = v_\xi |l\vec{0}\rangle\langle l\vec{0}| \tag{12}$$

where $v_\xi = V_0 I - \xi V_{ex}\sigma_z$, and $\xi = +1$ (-1) for the impurity's spin pointing up (down). Here we have assumed, without loss of generality, that the impurity occupies the site $\vec{R} = \vec{0}$ of plane l.

The change in the thermodynamic potential due to the introduction of the impurity in the system is now given by

$$\delta\Omega(\theta,\xi) = \frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Tr}\int_{-\infty}^{+\infty} \mathrm{d}\omega\; f(\omega)\ln\left[1 - G^{(0)}_{l,\vec{0};l,\vec{0}}(\omega,\theta)v_{\xi}\right]. \tag{13}$$

When we allow the impurity magnetic moment to point either up or down in both the ferromagnetic and antiferromagnetic configurations of the magnetic layers, we first calculate

$$\delta\Omega(0) = \min\{\delta\Omega(0,+1);\ \delta\Omega(0,-1)\} \tag{14}$$

and

$$\delta\Omega(\pi) = \min\{\delta\Omega(\pi,+1);\ \delta\Omega(\pi,-1)\}. \tag{15}$$

The impurity contribution to the coupling is then given by

$$\delta J = \delta\Omega(\pi) - \delta\Omega(0). \tag{16}$$

When the impurity is situated next to the magnetic plane (0), in the spacer, we find that its magnetic moment remains pinned to $\hat{m}_{\mathrm{A}}$ no matter what the orientation of $\vec{m}_{\mathrm{B}}$ is, which fully justifies the assumption that we made in subsection 2.1.

2.3. Magnetic plateaus at the interface

We now calculate the effects on the coupling due to the presence of a plateau of magnetic atoms at the interface between the magnetic plane (0) and the spacer. We consider p magnetic atoms occupying neighbouring sites on plane $l = 1$ of the spacer. As in the case of a single impurity at the interface, we assume that the magnetic moments of the atoms in the plateau are parallel to the magnetization of plane (0). The perturbing potential is given by

$$\hat{V} = v\sum_{s=1}^{p}|1,\vec{R}_s\rangle\langle 1,\vec{R}_s|. \tag{17}$$

The calculation of δJ is similar to that for the single impurity at the interface, the difference being that in the present case one has to deal with $(p \times p)$ matrices. After some manipulation we end up with the following expression:

$$\delta J = \frac{1}{\pi}\,\mathrm{Im}\int_{-\infty}^{+\infty}\mathrm{d}\omega\; f(\omega)\,\mathrm{Tr}\ln\left\{\left[\Lambda(\omega,\pi)\right]\left[\Lambda^{-1}(\omega,0)\right]\right\}$$

where

$$[\Lambda(\omega,\theta)] = [1] - [G^{(0)}(\omega,\theta)]v$$

with

$$[G^{(0)}(\omega,\theta)]_{(1,\vec{R}),(1,\vec{R}')} = \langle 1,\vec{R}|G^{(0)}(\omega,\theta)|1,\vec{R}'\rangle.$$

3. Results and conclusions

Here we present results on δJ for all of the situations considered in the previous section. In our calculations we have chosen the unit of energy such that the nearest-neighbour hopping $t = 1/2$. A typical value of $V_{ex} = 0.2$ was used, and for simplicity the non-magnetic part of the potential was taken as constant throughout the system, i.e., $V_0 = 0$. The effect of V_0 on the exchange coupling has been thoroughly investigated by Ferreira *et al* [17]. We have also assumed that the atomic planes have (0, 0, 1) orientation, and calculations were performed for

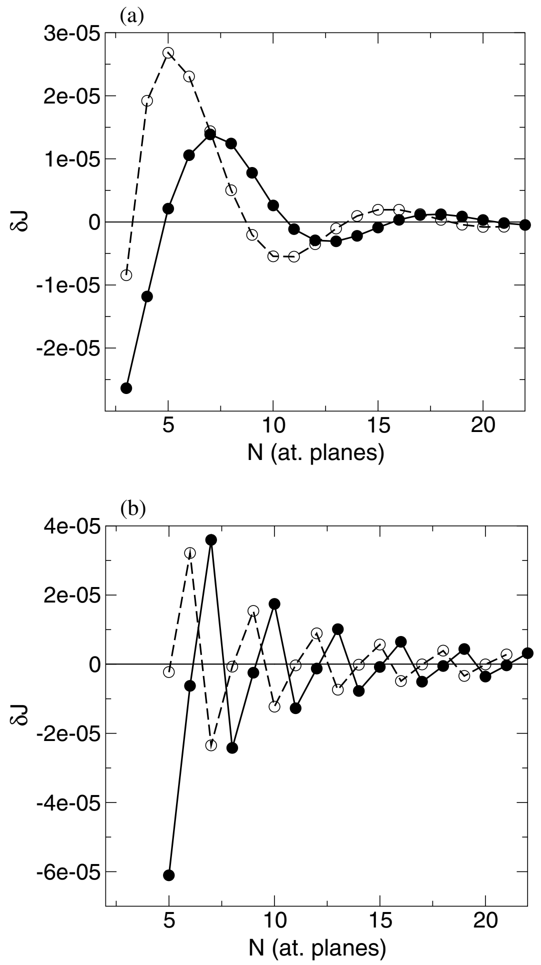

Figure 2. $\delta J(\pi)$ and $J^0(\pi)/N_\|$ (dashed line) for the impurity with spin up close to the first interface, for $E_F = -1.05$ (a) and $E_F = 2.5$ (b).

two different values of E_F, namely, -1.05 and 2.5, corresponding to long- and short-period oscillations of $J^{(0)}$, respectively.

Figure 2 shows δJ as a function of the spacer thickness for an impurity sitting next to magnetic plane (0), and for E_F equal to -1.05 (a) and 2.5 (b). For comparison, corresponding results for the coupling $J^{(0)}/N_\|$, where $N_\|$ is the number of atoms within each atomic plane, are also presented (open symbols). We notice that δJ oscillates as a function of the spacer thickness with the same period of J. This can be proved analytically in the asymptotic regime ($N \gg 1$), using the stationary-phase method [23–25]. The procedure is very close to that used to investigate the asymptotic behaviour of the coupling as a function of the spacer thickness. In both cases we find that the oscillation periods are determined by the extremal dimensions of the spacer Fermi surface in the direction perpendicular to the layers [5, 7, 24]. We notice that the amplitude of δJ is comparable to (or even greater than) that of $J^{(0)}/N_\|$. In addition, there is a phase difference between the two curves, which leads to a reduction of the coupling in the presence of magnetic impurities at the interface. Such an effect is more pronounced in

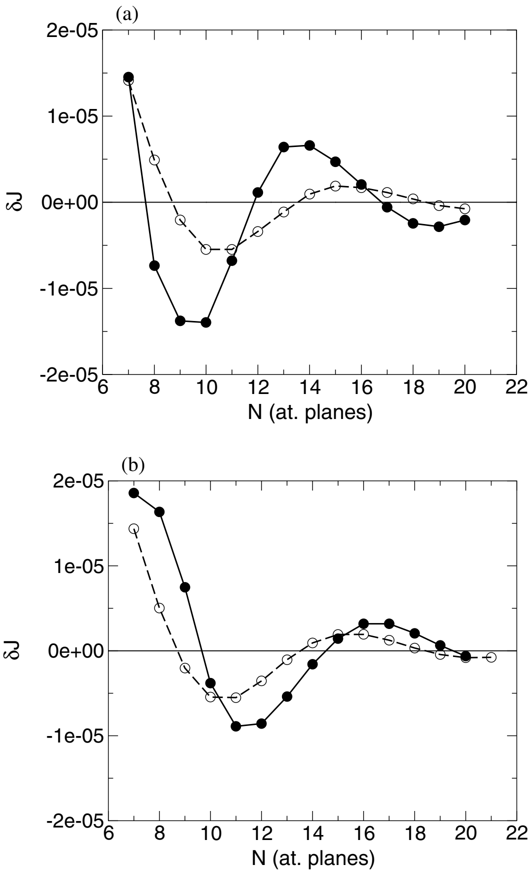

Figure 3. Weighted averages of $\delta J(\pi)$ over various impurity positions in the spacer for probability distributions P_1 (a) and P_2 (b) (see equation (18)). The calculated results are for $E_F = -1.05$. The dashed lines represent $J^{(0)}(\pi)/N_\parallel$.

the case of short-period oscillations (cf. figure 2(b)).

In order to assess the effects of magnetic impurities placed anywhere within the spacer layer, we have taken the average of δJ over the impurity position assuming two different probability distributions $P(l)$ for the impuity position, namely

$$P_1(l) = \frac{1}{N}$$
$$P_2(l) = \frac{1}{\mathcal{P}}(\mathrm{e}^{-l/\lambda} + \mathrm{e}^{(l-N+1)/\lambda}) \tag{18}$$

where

$$\mathcal{P} = \sum_{l=1}^{N} P_2(l).$$

In the first case, the impurity may occupy any atomic plane in the spacer with equal probability, whereas in the second it is more likely to be found close to one of the magnetic layers. The actual value of the parameter λ, which is determined by the impurity diffusion within the spacer, depends on the growth conditions and heat treatment that the sample might have been subjected to. In our calculations we have taken $\lambda = 0.2$.

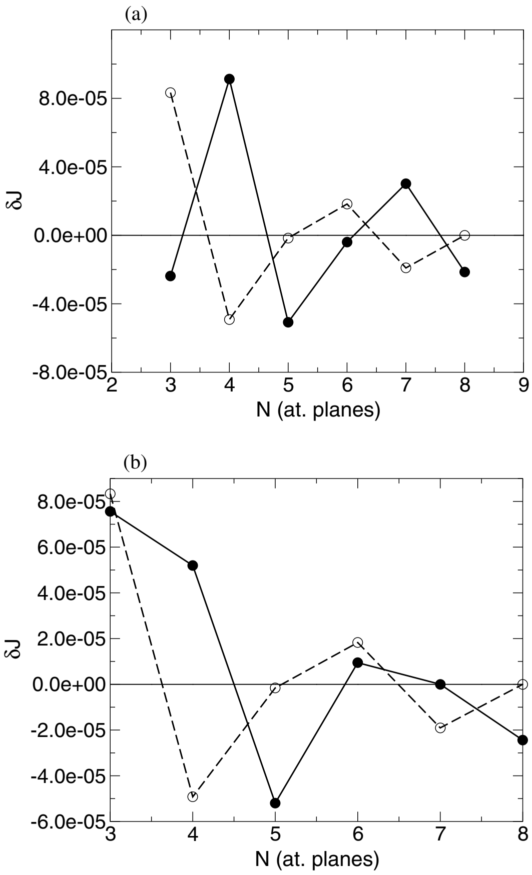

Figure 4. Weighted averages of $\delta J(\pi)$ over various impurity positions in the spacer for probability distributions P_1 (a) and P_2 (b) (see equation (18)). The calculated results are for $E_F = 2.5$. The dashed lines represent $J^{(0)}(\pi)/N_{\|}$.

Figure 3 depicts δJ (full symbols) as a function of the spacer thickness for $E_F = -1.05$, and distributions $P_1(l)$ (a) and $P_2(l)$ (b). In each case, open symbols correspond to $J^{(0)}/N_{\|}$. We notice that for long-period oscillations the effect on the coupling due to magnetic impurities is more pronounced when they are uniformly distributed within the spacer. However, for short-period oscillations, we find that δJ is not very sensitive to the impurity distribution. This is made clear in figure 4, which shows the change in the coupling for $E_F = 2.5$ and for the two distributions $P_1(l)$ (a) and $P_2(l)$ (b). Here again open symbols represent $J^{(0)}/N_{\|}$.

Now we turn our attention to the case of magnetic plateaus at the interface of the magnetic plane (0) and the spacer. This is a very interesting situation, since the calculation of δJ now takes into account the effects of quantum interference between electronic waves scattered by neighbouring atoms in the plateau. In figure 5 we show the results for δJ for a two-atom plateau (full circles) and a four-atom plateau (full squares), for $E_F = -1.05$. Corresponding results for $E_F = 2.5$ are presented in figure 6. For the two Fermi energies, we notice that to a good approximation δJ scales with the number of atoms in the plateau. This is particularly clear in the case of short-period oscillations ($E_F = 2.5$), as shown in figure 7, where δJ for $N = 7$ is presented as a function of the number of magnetic atoms in the plateau, for both

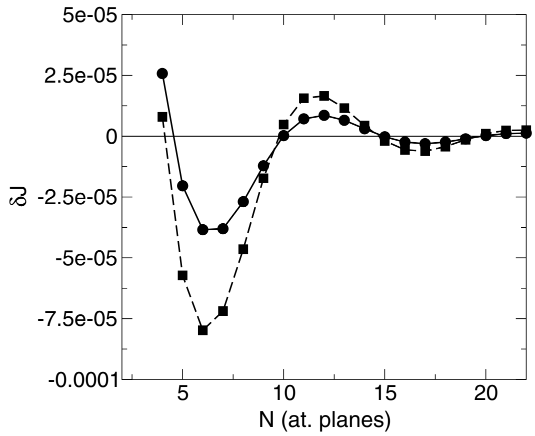

Figure 5. Exchange-coupling variation $\delta J(N)$ in the presence of two-impurity (solid line) and four-impurity (dashed line) clusters in the first plane of the non-magnetic spacer, for Fermi energy $E_F = -1.05$.

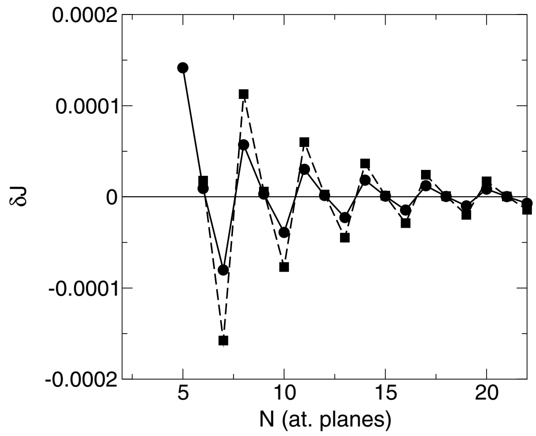

Figure 6. Exchange-coupling variation $\delta J(N)$ in the presence of two-impurity (solid line) and four-impurity (dashed line) clusters in the first plane of the non-magnetic spacer, for Fermi energy $E_F = 2.5$.

$E_F = -1.05$ (full squares) and $E_F = 2.5$ (full circles). On the basis of these results, one might obtain a reasonable approximation for δJ in the case of a finite concentration x of impurities by simply multiplying δJ by x, at least in the dilute impurity regime. Such a result supports the use of the coherent potential approximation to calculate the interlayer coupling.

In conclusion, we have investigated the effects of imperfections in the crystalline structure on the exchange coupling in metallic multilayers. In the case of magnetic impurities in the spacer, we have found that for long-period oscillations their effect on the coupling is more pronounced when they are uniformly distributed. On the other hand, for short-period oscillations, δJ does not turn out to be very sensitive to the impurity distribution. Our results indicate that the effects of a finite concentration of impurities may be superimposed, at least in the low-concentration regime.

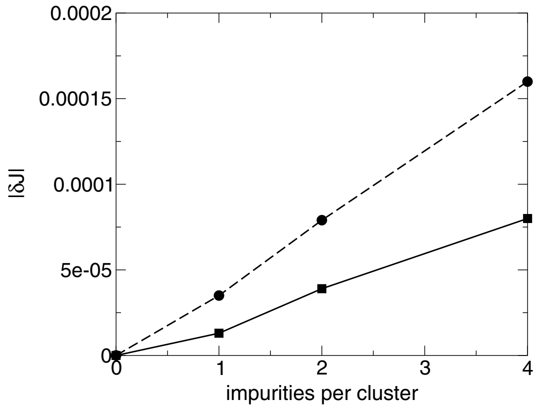

Figure 7. The amplitude $|\delta J|$ for varying cluster size for Fermi energies $E_F = -1.05$ (solid line) and $E_F = 2.5$ (dashed line). A strong growth of the impurity's contribution with the size of the clusters is clearly seen.

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