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2002 J. Phys.: Condens. Matter 14 4355

(http://iopscience.iop.org/0953-8984/14/17/308)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) 4355–4363 PII: S0953-8984(02)30764-1

Interlayer exchange coupling in M/N/M multilayers

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Received 8 November 2001, in final form 8 March 2002
Published 18 April 2002
Online at stacks.iop.org/JPhysCM/14/4355

Abstract

The interlayer exchange coupling (IEC) of two local moment ferromagnetic layers separated by a non-magnetic spacer layer (a M/N/M multilayer) is studied using the modified Ruderman–Kittel–Kasuya–Yosida (RKKY) method along with the s–f model. The IEC exhibits oscillatory behaviour with respect to the spacer layer thickness and it oscillates between ferromagnetic and antiferromagnetic configurations. The conventional RKKY method is also used to obtain the IEC and the results are compared with those obtained from the modified RKKY method which incorporates the electron correlation effects. We find significant correlation effects on the IEC and in fact the correlations alter the nature and magnitude of the magnetic coupling. Hence this study indicates the importance of the inclusion of correlation effects to the understanding of the IEC in multilayer systems with local moment ferromagnetic sublayers and to offering a satisfactory explanation for the experimental results.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The indirect exchange interaction between two ferromagnetic layers separated by a non-magnetic spacer layer exhibits oscillatory behaviour with respect to the spacer layer thickness and this has been observed experimentally in many magnetic multilayer systems [1–6]. The interpretations of these experimental observations are provided by methods based on the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction [7,8], the Hubbard-type model [9], the s–f exchange model [10] and *ab initio* total-energy calculations [11–13]. Naive application of the RKKY method (assuming a spherical Fermi surface and uniform distribution of the spins in the ferromagnetic layers) leads to small oscillation periods [7]. Hence Bruno and Chappert [8] extended the general theory of the RKKY method and got satisfactory oscillation periods for Co/Cu/Co and Fe/Cu/Fe multilayers considering the topological properties of the Fermi surface of the spacer layer and the moment distribution within the ferromagnetic layers. Using a Hubbard-like one-band model [9], Edwards *et al* showed that the exchange coupling

exhibits long-period oscillations with respect to the spacer layer thickness for certain positions of the Fermi level. Urbaniak-Kucharczyk used the s–f exchange model to study the interlayer coupling, expressing the effective exchange integrals in terms of the electron susceptibility [10]. Convincing results for the fcc (111), (100) and (110) spacers are found. *Ab initio* total-energy methods provide a rather straightforward approach to the problem. However, they are not adequate for strongly correlated systems. An extensive discussion of the general theory of the interlayer exchange is given by Bruno [14] and an illustration of the theory for the case of Co/Cu/Co multilayers is also presented by him. The interlayer exchange coupling (IEC) is found to have significant influence on the magnetic properties of the ferromagnetic sublayers. Ney *et al* [1] found an oscillating behaviour in T_c for Co/Cu/Ni trilayers with respect to the spacer layer thickness and a theoretical analysis of this effect based on the Hubbard model is reported by Wu *et al* [15].

Our interest is in studying the exchange coupling of the ferromagnetic (M) layers in M/N/M multilayers with M being a local moment ferromagnetic metal and N being a non-magnetic metal. In the local moment metals, the magnetic properties are dominated by the intra-atomic exchange interaction acting between the conduction electrons and the local moments (s–f exchange) and the magnetic coupling between the local moments situated at the lattice sites is mediated by the conduction electrons. The above-mentioned correlation effects are properly taken into account in the s–f model, which is often referred to as the ferromagnetic Kondo-lattice model in the recent literature, and the modified RKKY method [16, 17] describes the exchange coupling of the localized spins mediated by the correlated conduction electrons. Thus this approach leads to the self-consistent evaluation of the exchange integrals acting between the local moments situated at the lattice sites and hence we use this s–f model along with the modified RKKY method to study the IEC in M/N/M multilayers with M being a local moment metal. The starting Hamiltonians of our method and the method of Urbaniak-Kucharczyk [10] are similar but the approaches are completely different (see section 5). Further, our method and Urbaniak-Kucharczyk's method are different from Bruno's method [14] in the sense that in Bruno's method, the IEC is derived from the interference effects of the electron waves whereas in our method and Urbaniak-Kucharczyk's method, the IEC is derived using the indirect coupling of the localized spins mediated by the conduction electrons. In our method the full Green function of the s–f system is evaluated and the IEC is studied using the above-mentioned modified RKKY method. In the present work, we have carried out our study for a model system with only one conduction band per layer and we hope that this study will form the basis for a calculation for a real system. The influence of the IEC on the magnetic properties such as the magnetization and Curie temperature of the ferromagnetic sublayer can also be studied using this method. However, this study is planned for a future paper. In this paper we present our results on the IEC in the M/N/M multilayer at $T = 0$ K with different thicknesses of the non-magnetic spacer layer. We have further studied the influence of the conduction electron concentration on the IEC. We present our results in the following sections along with a brief discussion of the s–f model and the modified RKKY method.

2. s–f model for the ferromagnetic films

In this section we shall present a brief discussion of the s–f model. We shall present the theory for an n-layer film and the theory can then easily be adapted to the required M/N/M multilayer geometry by assuming that there are no localized spins in the non-magnetic spacer layers. The theory and the mathematical formulation of the s–f model are described in many of the earlier publications [16, 17]. Hence we will not present those details here. However, for the sake of completeness, we will present a brief summary of the model. We will consider a ferromagnetic

film with n layers. The film is characterized by a two-dimensional Bravais lattice vector having an n-atom basis. The n-atom basis corresponds to the n layers of the film. A lattice vector of the film may be given as

$$\boldsymbol{R}_{i\alpha} = \boldsymbol{R}_i + \boldsymbol{r}_\alpha.$$

$\boldsymbol{R}_i$ is the two-dimensional Bravais lattice vector and $\boldsymbol{r}_\alpha$ is the basis vector. At each site of the film a localized spin S is present and the film is assumed to have only one conduction band per layer. The electrons in the conduction band are exchange coupled to the local moments. This situation is well described by the s–f model, the model Hamiltonian of which is given as

$$H = \sum_{\substack{ij,\sigma \\ \alpha\beta}} t_{ij}^{\alpha\beta} c_{i\alpha\sigma}^{\dagger} c_{j\beta\sigma} - j \sum_{j,\alpha} \boldsymbol{S}_{j\alpha} \cdot \boldsymbol{\sigma}_{j\alpha}.$$

The first term describes the conduction electrons and the second term represents the interaction of the conduction electrons with the local moments. i, j represent the site indices of the two-dimensional lattice and α, β represent the layer indices. $t_{ij}^{\alpha\beta}$ is the hopping integral and j is the s–f exchange coupling strength. All the information concerning the electronic structure and magnetic properties of the system described by the above Hamiltonian can be obtained from the retarded single-electron Green function

$$G_{ij\sigma}^{\alpha\beta} = \langle\langle c_{i\alpha\sigma}; c_{j\beta\sigma}^{\dagger} \rangle\rangle_E = -\mathrm{i} \int_0^\infty \mathrm{d}t\, \mathrm{e}^{(\mathrm{i}/\hbar)Et} \langle [c_{i\alpha\sigma}(t), c_{j\beta\sigma}^{\dagger}(0)]_+ \rangle.$$

Evaluation of this Green function proceeds by the equation-of-motion method. The equation of motion is written as

$$\sum_{r\gamma} (E\delta_{ir} - T_{ir}^{\alpha\gamma}) G_{rj\sigma}^{\gamma\beta}(E) = \hbar\delta_{ij}\delta_{\alpha\beta} + \langle\langle [c_{i\alpha\sigma}, H_{sf}]_-; c_{j\beta\sigma}^{\dagger} \rangle\rangle_E.$$

Defining the self-energy $M_{ij\sigma}(E)$ as

$$\langle\langle [c_{i\alpha\sigma}, H_{sf}]_-; c_{j\beta\sigma}^{\dagger} \rangle\rangle_E = \sum_{r\gamma} M_{ir\sigma}^{\alpha\gamma}(E) G_{rj\sigma}^{\gamma\beta}(E)$$

and making a Fourier transformation (with respect to the spatial variables) of the equation of motion leads to

$$\sum_{\gamma} (E\delta_{\alpha\gamma} - \varepsilon_{\alpha\gamma}(\boldsymbol{k}) - M_{\boldsymbol{k}\sigma}^{\alpha\gamma}(E)) G_{\boldsymbol{k}\sigma}^{\gamma\beta}(E) = \hbar\delta_{\alpha\beta}.$$

In the matrix form the above equation may be written as

$$\hat{G}_{\boldsymbol{k}\sigma}(E) = \frac{\hbar}{[E\hat{I} - \hat{\varepsilon}(\boldsymbol{k}) - \hat{M}_{\boldsymbol{k}\sigma}(E)]}.$$

$\hat{\varepsilon}(\boldsymbol{k})$ is the Bloch energy matrix and for an n-layer film it will take the form

$$\hat{\varepsilon}(\boldsymbol{k}) = \begin{pmatrix} \varepsilon^{11}(\boldsymbol{k}) & \varepsilon^{12}(\boldsymbol{k}) & \cdots & \varepsilon^{1n}(\boldsymbol{k}) \\ \varepsilon^{21}(\boldsymbol{k}) & \varepsilon^{22}(\boldsymbol{k}) & \cdots & \varepsilon^{2n}(\boldsymbol{k}) \\ \vdots & \vdots & \vdots & \vdots \\ \varepsilon^{n1}(\boldsymbol{k}) & \varepsilon^{n2}(\boldsymbol{k}) & \cdots & \varepsilon^{nn}(\boldsymbol{k}) \end{pmatrix}.$$

The diagonal elements are the intralayer Bloch energies and the off-diagonal elements are the interlayer Bloch energies.

$\hat{M}_\sigma(E)$ is the self-energy matrix and it takes the form

$$\hat{M}_\sigma(E) = \begin{pmatrix} M_\sigma^{11}(E) & M_\sigma^{12}(E) & \cdots & M_\sigma^{1n}(E) \\ M_\sigma^{21}(E) & M_\sigma^{22}(E) & \cdots & M_\sigma^{2n}(E) \\ \vdots & \vdots & \vdots & \vdots \\ M_\sigma^{n1}(E) & M_\sigma^{n2}(E) & \cdots & M_\sigma^{nn}(E) \end{pmatrix}.$$

The diagonal elements are the intralayer self-energies and the off-diagonal elements are the interlayer self-energies.

The layer-dependent spectral density may now be obtained from the Green function matrix as

$$S^{\alpha\alpha}_{k\sigma}(E) = -\frac{1}{\pi}\,\mathrm{Im}\,G^{\alpha\alpha}_{k\sigma}(E).$$

The layer-dependent quasiparticle density of states (QDOS) is

$$\rho^{\alpha\alpha}_{\sigma}(E) = \sum_{k} S^{\alpha\alpha}_{k\sigma}(E).$$

The evaluation of the self-energy is described in many of the earlier publications [18–20]. The general form of the intralayer self-energy is

$$M_{\sigma}(E) = -\tfrac{1}{2} j z_{\sigma} \langle S^{Z} \rangle + \tfrac{1}{4} j^{2} K_{\sigma}(E).$$

The first term, which is exact in the weak-coupling limit, represents an induced 'Slater splitting' of the band proportional to the f-spin magnetization $\langle S^{Z} \rangle$. The second term is dominated by the consequences of the process of spin exchange between the conduction electrons and the local moments and hence $K_{\sigma}(E)$ turns out to be a complicated functional of the self-energy for both spin directions. In addition, $K_{\sigma}(E)$ involves several pure and mixed-spin correlation functions too [16, 18]. Thus the self-energy is to be calculated self-consistently—along with the self-consistent evaluation of the spin correlation functions. The interlayer self-energy is negligible in the case of the M/N/M multilayer and hence it is not considered in our calculations. Once the self-energy is calculated, the matrix Green function of the film can immediately be obtained.

It should be mentioned that the approximation used in [16] and [18] does not obey the Luttinger theorem, i.e. there is in general a finite imaginary part of the self-energy at the Fermi edge. We believe that this is a shortcoming of the sophisticated approximation scheme rather than a hint of a real non-Fermi-liquid behaviour. On the other hand, in our opinion it remains an interesting unsolved problem whether or not we indeed have to expect Fermi-liquid behaviour in the s–f model (the ferromagnetic Kondo-lattice model), which does not describe a purely fermionic system (localized spins!) The various spin-flip-exchange processes (magnon emission and absorption scattering states, magnetic polarons) may destroy the picture.

3. Modified RKKY method

The modified RKKY method essentially aims at the evaluation of the indirect exchange coupling between two localized moments mediated by the conduction electrons which are coupled to the localized spins situated at the lattice sites through the s–f exchange acting between them and the localized spins. In order to obtain this effective exchange interaction between the localized spins, the s–f interaction is mapped to an effective Heisenberg Hamiltonian by averaging out the conduction electron degrees of freedom [17]. This procedure allows us to introduce an effective exchange coupling between the localized moments as a functional of the conduction electron self-energy. The details of the method may again be found in the earlier publications [17] and hence we will not elaborate the method here. However, for the sake of completeness, we will quote here the required results which are used in the calculations. The exchange integral acting between two localized moments is given by this method as

$$J^{\alpha\beta}_{ij} = \frac{1}{N}\sum_{\boldsymbol{q}} J^{\alpha\beta}(\boldsymbol{q}) \mathrm{e}^{-\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}_{i\alpha}-\boldsymbol{R}_{j\beta})}$$

where $J^{\alpha\beta}(\boldsymbol{q})$ is given as

$$J^{\alpha\beta}(\boldsymbol{q}) = \frac{1}{8\pi} j^2 \sum_{\sigma} \mathrm{Im} \int_{-\infty}^{\infty} \mathrm{d}E \; f_-(E) \frac{1}{N\hbar} \sum_{\boldsymbol{k}} A^{\sigma\sigma,\alpha\beta}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}(E)$$

and

$$\hbar A^{\sigma\sigma,\alpha\beta}_{\boldsymbol{k},\boldsymbol{k}+\boldsymbol{q}}(E) = (\hat{G}^{(0)}_{\boldsymbol{k}}(E)\hat{G}_{\boldsymbol{k}+\boldsymbol{q}\sigma}(E))^{\alpha\beta} + (\hat{G}^{(0)}_{\boldsymbol{k}+\boldsymbol{q}}(E)\hat{G}_{\boldsymbol{k}\sigma}(E))^{\alpha\beta}.$$

$\hat{G}^{(0)\alpha\beta}_{\boldsymbol{k}}$ is the free-electron Green function and $\hat{G}^{\alpha\beta}_{\boldsymbol{k}\sigma}(E)$ is the already defined Green function of the s–f system. $f_-(E)$ is the Fermi function. The $\boldsymbol{q}$-dependent exchange integrals can thus also be written in a matrix form for an n-layer film as

$$\hat{J}(\boldsymbol{q}) = \begin{pmatrix} J^{11}(\boldsymbol{q}) & J^{12}(\boldsymbol{q}) & \cdots & J^{1n}(\boldsymbol{q}) \\ J^{21}(\boldsymbol{q}) & J^{22}(\boldsymbol{q}) & \cdots & J^{2n}(\boldsymbol{q}) \\ \vdots & \vdots & \vdots & \vdots \\ J^{n1}(\boldsymbol{q}) & J^{n2}(\boldsymbol{q}) & \cdots & J^{nn}(\boldsymbol{q}) \end{pmatrix}.$$

As the exchange integrals are dependent on the Green function of the s–f system, it is obvious that they are dependent on the electron self-energy.

It is possible to obtain the conventional RKKY interaction from this formalism through the first-order approximation

$$G^{\alpha\beta}_{\boldsymbol{k}\sigma}(E) \to G^{(0)\alpha\beta}_{\boldsymbol{k}\sigma}(E)$$
$$G^{\alpha\beta}_{\boldsymbol{k}+\boldsymbol{q}\sigma}(E) \to G^{(0)\alpha\beta}_{\boldsymbol{k}+\boldsymbol{q}\sigma}(E).$$

In this case $J^{\alpha\beta}(\boldsymbol{q})$ will be identical to the well-known RKKY expression.

4. Interlayer exchange coupling

Our primary interest is in evaluating the indirect exchange coupling between two local moment ferromagnetic layers separated by a non-magnetic spacer layer including the effects of the above-mentioned s–f exchange acting between the conduction electrons and the localized spins of the ferromagnetic layers. Hence we have made use of the modified RKKY method to evaluate the required exchange integrals. The matrix Green function of the n-layer film can easily be converted to describe the M/N/M multilayer by assuming that there are no localized spins in the spacer layers and hence the Green functions of the spacer layers will be just the free-electron Green functions. The matrix Green function of the M/N/M multilayer can thus be generated and it can subsequently be used to obtain the exchange integral matrix $\hat{J}(\boldsymbol{q})$ discussed in the previous section. The exchange interaction acting between two moments located in the bottom and topmost ferromagnetic sublayers (J_{ij}) of the multilayer can now be calculated from $\hat{J}(\boldsymbol{q})$. Summation over all the exchange integrals (J_{ij}) acting between the localized moments situated at the lattice sites of these two ferromagnetic layers will yield the IEC. In all our calculations, we assumed the geometry of two ferromagnetic monolayers separated by a spacer layer and the spacer layer thickness was varied by up to thirty monolayers. The geometry of the multilayer with a spacer thickness of five monolayers is shown in figure 1. The Bravais lattice of each layer is assumed to be a square lattice and the nearest-neighbour intralayer and interlayer electron hopping is assumed to occur in the multilayer. The magnitude of the hopping is assumed to be the same in all the layers. The value of the s–f coupling constant j is assumed to be 0.2 eV and the magnitude of the localized spin is taken as $7/2$. The electron self-energy is actually dependent on the band occupation and temperature, and hence the exchange integrals will also be dependent on the band occupation and temperature. We

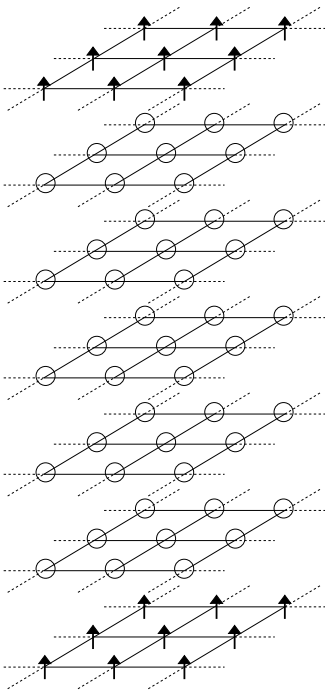

Figure 1. The geometry of the M/N/M multilayer with a spacer thickness of five monolayers.

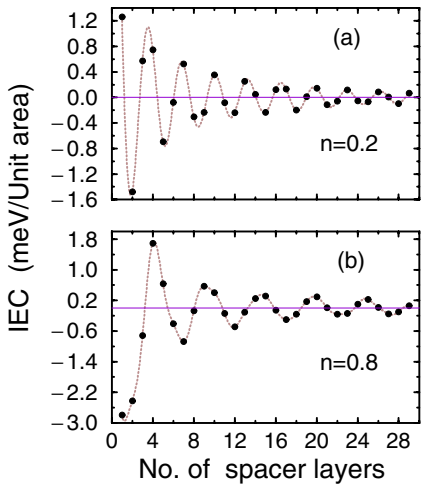

Figure 2. The IEC obtained from the modified RKKY method at various values of the spacer thickness for two different band occupations ($T = 0$ K).

have studied the influence of the band occupation on the exchange integrals, keeping the same band occupation in all the layers, and a study of the influence of temperature on the IEC is planned for a forthcoming paper. We have further evaluated the interlayer coupling using the conventional RKKY method and the results of our calculations are discussed in the following section.

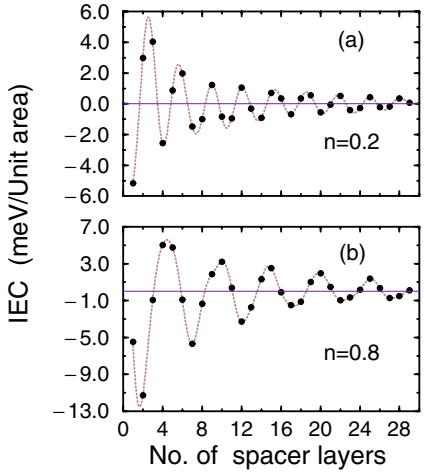

Figure 3. The IEC obtained from the conventional RKKY method at various values of the spacer thickness for two different band occupations ($T = 0$ K).

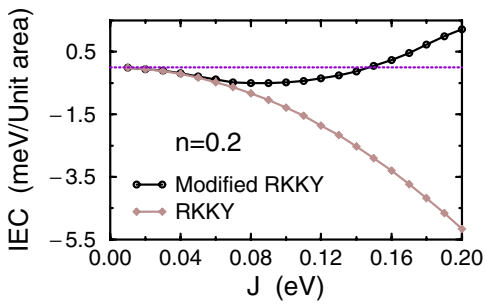

Figure 4. The variation of the IEC with respect to the s–f exchange coupling strength (j) for the M/N/M multilayer with a monolayer spacer ($T = 0$ K).

5. Discussion of results

The IEC calculated using the modified RKKY method at two different band occupations ($n = 0.2, 0.8$) are presented in figure 2. The IEC oscillates between ferromagnetic and antiferromagnetic configurations. This behaviour is in agreement with that of the trend seen in many experimental works [2, 4]. As it is a model calculation, the oscillation periods and nature of the magnetic coupling with respect to the spacer layer thickness cannot be compared quantitatively with the experimental works. Hence our next goal is to extend the calculations to a real material film where the calculational complexity however will be enormous. Our interest in this study is in indicating the importance of electron correlation effects (self-energy) in deciding the nature of the magnetic coupling of the ferromagnetic layers with respect to the spacer layer thickness and in influencing the oscillation periods. Hence we carried out the calculations using the conventional RKKY method also. The results obtained using the conventional RKKY method for the same band occupations are presented in figure 3. It may be seen from the figures that the influence of correlation on the magnetic coupling is significant. When the spacer consists of one monolayer, the coupling is antiferromagnetic for the uncorrelated film (figure 3(a)) whereas it is ferromagnetic for the correlated film (figure 2(a)). The behaviours of the magnetic coupling at larger spacer layer thicknesses also

show significant differences between the uncorrelated and correlated films. The magnitude of the coupling strength is also drastically modified because of the correlation effects and this may also be seen from the figures. Thus our studies clearly demonstrates the influence of the correlation effects on the exchange coupling of the ferromagnetic layers. In order to study the evolution of the correlation effects on the interlayer coupling, we have further calculated the IEC as a function of the s–f exchange coupling strength (j) for the multilayer with a single spacer layer using the modified RKKY method as well as the conventional RKKY method. The results are plotted in figure 4. It may be seen from the figure that both the methods predict antiferromagnetic exchange coupling at small values of j. However, after a critical value of j, the modified RKKY method predicts a transition of the exchange coupling to a ferromagnetic nature, whereas the conventional RKKY method predicts that the antiferromagnetic coupling is retained for all values of j. The effect of correlation on the IEC is thus clearly demonstrated in figure 4. The modified RKKY method further predicts that the IEC gets saturated after a certain value of j, whereas in the case of conventional RKKY method, the IEC can never reach saturation. As the modified RKKY method uses the full Green function of the system which properly takes into account the s–f interaction, it is obvious that these calculations are much more reliable than the conventional RKKY method. On the other hand, in the method of Urbaniak-Kucharczyk the effective exchange integrals are expressed in terms of the electron susceptibility and the s–f interaction strength j. The expression for the effective exchange integrals is quadratic in j and hence the behaviour of the IEC seen in figure 4 is not obtainable from his method. The IEC is found to be sensitive to the band occupation also and this may be seen from figures 2 and 3. The band occupation also alters the nature and magnitude of the IEC. In order to obtain the oscillation periods, we performed a Fourier analysis of our data. The dotted lines shown in figures 2 and 3 are the Fourier fits to our data. The Fourier analysis of our data corresponding to the modified RKKY method revealed that the oscillation period at $n = 0.2$ is 3.22 whereas it is 4.83 when $n = 0.8$. Thus we found only a single-period oscillation in the IEC. The oscillation periods are almost the same for the RKKY data also. It may be seen from figures 2 and 3 that there is an overall sign change of the decaying wave from the modified RKKY method to conventional RKKY method for $n = 0.2$, whereas it is absent for $n = 0.8$. The magnitude of the oscillation is very much reduced in the modified RKKY method compared to that of the conventional RKKY method. This reduction in the magnitude of the oscillations is certainly due to the effects of correlation and our results might have been underestimated a little bit due to the presence of the finite imaginary part of the self-energy at the Fermi edge (Luttinger theorem). The oscillation period is found to depend on the band occupation. Our model considers only one conduction band per atomic layer and hence if there are N atomic layers in the spacer then there will be N bands. These N bands will contribute to the Fermi surface according to the band filling and the Fermi surface will obviously have many sheets. At a given band occupation the variations of the IEC and the Fermi surface with respect to the spacer layer thickness are related. This is the reason for getting different oscillation periods for $n = 0.2$ and 0.8 in our calculations. The analysis of our results shows that the asymptotic behaviour of the IEC at large spacer thickness exhibits approximately the d^{-2}-behaviour.

6. Conclusions

We have evaluated the IEC of two local moment ferromagnetic sublayers in a M/N/M multilayer at different spacer thicknesses. The calculations were carried out using the s–f model and the modified RKKY method which provide a self-consistent description of the exchange coupling between the moments situated at the lattice sites of the two ferromagnetic sublayers.

The IEC is found to have an oscillating behaviour with respect to the spacer layer thickness and it oscillates between ferromagnetic and antiferromagnetic configurations. In order to demonstrate the influence of correlation effects on the IEC, we have further evaluated it using the conventional RKKY method too. We find significant influences of the electron correlation effects on the IEC and the correlation effects are found to change the nature and magnitude of the exchange coupling. The variation of the IEC with respect to the band occupation is also studied using both the methods and the band occupation also found to alter the nature and magnitude of the IEC. The oscillation period of the IEC is also found to depend on the band occupation. Our method is very much more sophisticated than the conventional RKKY method and hence a calculation for real local moment multilayers can be expected give reliable and interesting results. It should be borne in mind that the generally occurring imperfections in the interfaces of the artificially prepared multilayers, such as the interfacial roughness and interfacial interdiffusion, may have to be considered when the results for real material multilayers are compared with the experimental results [21].

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