Interlayer exchange coupling in M/N/M multilayer

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The interlayer exchange coupling (IEC) of two local moment ferromagnetic layers separated by a non-magnetic spacer layer (M/N/M multilayer) is studied using the modified RKKY method along with the s-f model. The IEC exhibits oscillatory behaviour with respect to the spacer layer thickness and it oscillates between ferro- 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 points out the importance of the inclusion of correlation effects in the understanding of the IEC in multilayer systems with local moment ferromagnetic sub-layers and to offer satisfactory explanation for the experimental results.

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 interpretation of these experimental observations are provided by methods based on the RKKY interaction [7,8], Hubbard type model [9], s-f exchange model [10] and ab-initio total energy calculations [11-13]. Naive application of the RKKY method (assuming spherical Fermi surface and uniform distribution of 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 are restricted to small spacer layer thickness as they become very difficult when the spacer thickness is large. An extensive discussion of the general theory of the interlayer exchange is given by Bruno [14] and the illustration of the theory for the case of Co/Cu/Co trilayer is also presented by him. The interlayer exchange coupling is found to have significant influence on the magnetic properties of the ferromagnetic sublayers. Ney et. al. found an oscillating behaviour in the $T_c$ of Co/Cu/Ni trilayers with respect to the spacer layer thickness [1] and a theoretical analysis of this effect based on the Hubbard model is reported by Wu et.al. [15].

Our interest is to study 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 nonmagnetic 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 localised 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 interlayer exchange coupling 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 sect.V). Further, our method and the Urbaniak-Kucharczyk’s method are different from the Bruno’s method [14] in the sense that in the Bruno’s method, the IEC is derived from the interference effects of the electron waves whereas in our method and the Urbaniak-Kucharczyk’s method, the IEC is derived using the indirect coupling of the localised spins mediated by the conduction electrons. In our method the full Green function of the $s – f$ system is evaluated and the interlayer exchange coupling (IEC) is studied using the above said 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 the calculation for a real system. The influence of the interlayer exchange coupling on the magnetic properties 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 interlayer exchange coupling in the M/N/M multilayer at T=0K 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.

II. 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 for the required M/N/M multilayer geometry by assuming that there are no localised 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 completion, we will present a brief summary of the model. We will consider a ferromagnetic film with n layers. The film is characterised 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

\[ \mathbf{R}_{i\alpha} = \mathbf{R}_i + \mathbf{r}_\alpha \]

\( \mathbf{R}_i \) is the two dimensional Bravais lattice vector and \( \mathbf{r}_\alpha \) is the basis vector. At each site of the film a localised 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

\[ H = \sum_{i,\alpha,\beta} t_{ij}^{\alpha\beta} c_{i,\alpha\sigma}^\dagger c_{j,\beta\sigma} - \sum_{j,\alpha} S_{j,\alpha \cdot \sigma} \cdot \sigma_{j,\alpha} \]

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 \( \alpha, \beta \) 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 said Hamiltonian can be obtained from the retarded single electron Green function

\[ G_{i\alpha\sigma}^{\alpha\beta}(E) = \langle c_{i,\alpha\sigma}(t), c_{j,\beta\sigma}^\dagger(0) \rangle_E \]

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

\[ \sum_{\alpha\gamma}(E \delta_{ir} - T_{ir}^{\alpha\gamma})G_{\gamma j\sigma}(E) = \hbar \delta_{ij} \delta_{\alpha\beta} + \langle c_{i,\alpha\sigma}, H_{sf} \rangle - \langle c_{i,\alpha\sigma}\rangle \langle c_{j,\beta\sigma} \rangle \]

Defining the selfenergy \( M_{i\alpha\sigma} \) as

\[ \langle c_{i,\alpha\sigma}, H_{sf} \rangle - \langle c_{i,\alpha\sigma}\rangle \langle c_{j,\beta\sigma} \rangle \]

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

\[ \sum_{\gamma}(E \delta_{ir} - \varepsilon_{r\gamma}(k) - M_{k\sigma}^{\alpha\gamma}(E))G_{\gamma j\sigma}(E) = \hbar \delta_{ij} \delta_{\alpha\beta} \]

In the matrix form the above equation may be written as

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

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

\[ \hat{\varepsilon}(k) = \begin{pmatrix} \varepsilon_{11}(k) & \varepsilon_{12}(k) & \cdots & \varepsilon_{1n}(k) \\ \varepsilon_{21}(k) & \varepsilon_{22}(k) & \cdots & \varepsilon_{2n}(k) \\ \vdots & \vdots & \ddots & \vdots \\ \varepsilon_{n1}(k) & \varepsilon_{n2}(k) & \cdots & \varepsilon_{nn}(k) \end{pmatrix} \]

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

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

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

\[ S_{k\sigma}^{\alpha\sigma}(E) = -\frac{1}{\pi} ImG_{k\sigma}^{\alpha\sigma}(E) \]
The layer dependent quasiparticle density of states (QDOS) is
\[ \rho_\sigma^{\alpha}(E) = \sum_k S_{k\sigma}^{\alpha}(E) \]

The evaluation of the self-energy is described in many of the earlier publications [18-20]. Once the self-energy is calculated, the matrix Green function of the film can immediately be obtained.

### III. MODIFIED RKKY METHOD

The modified RKKY method essentially aims at the evaluation of the indirect exchange coupling between two localised moments mediated by the conduction electrons which are coupled to the localised spins situated at the lattice sites through the s – f exchange acting between them and the localised spins. In order to obtain this effective exchange interaction between the localised 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 localised 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 completion, we will quote here the required results which are used in the calculations. The exchange integral acting between two localised moments is given by this method as

\[ J_{ij}^{\alpha\beta} = \sum_q J^{\alpha\beta}(q) e^{-i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)} \]

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

\[ J^{\alpha\beta}(q) = \frac{1}{8\pi^2} \int_q \sum_\sigma \frac{1}{E f_-(E)} \frac{1}{N_h} \sum_k A_{k,k+q}^{\sigma,\alpha\beta}(E) \]

and

\[ h A_{k,k+q}^{\sigma,\alpha\beta}(E) = (\hat{G}_{k}^{(0)}(E)\hat{G}_{k+q\sigma}(E))^{\alpha\beta}+(\hat{G}_{k+q}^{(0)}(E)\hat{G}_{k\sigma}(E))^{\alpha\beta} \]

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

\[ G_{k\sigma}^{\alpha\beta}(E) \to G_{k\sigma}^{(0)\alpha\beta}(E) \]

\[ G_{k+q\sigma}^{\alpha\beta}(E) \to G_{k+q\sigma}^{(0)\alpha\beta}(E) \]

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

### IV. INTERLAYER EXCHANGE COUPLING

Our primary interest is to evaluate the indirect exchange coupling between two local moment ferromagnetic layers separated by a nonmagnetic spacer layer including the effects of the above mentioned s – f exchange acting between the conduction electrons and the localised 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 localised 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 thus can be generated and it can subsequently be used to obtain the exchange integral matrix \( \hat{J}(q) \) discussed in the previous section. The exchange interaction acting between two moments located in the bottom and topmost ferromagnetic sub-layers (\( J_{ij} \)) of the multilayer can now be calculated from \( \hat{J}(q) \). Summation over all the exchange integrals \( (J_{ij}) \) acting between the localised moments situated at the lattice sites of these two ferromagnetic layers will yield the interlayer exchange coupling. In all our calculations, we assumed the geometry of two ferromagnetic monolayers separated by a spacer layer and the spacer layer thickness was varied up to thirty monolayers. The geometry of the multilayer with a spacer thickness of five monolayers is shown in Fig.1.

The Bravais lattice of each layer is assumed to be a square lattice and the nearest neighbor 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 values of the s – f coupling constant \( J \) is assumed to be 0.2 eV and the magnitude of the localised 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 have studied the influence of the band occupation on the exchange integrals keeping the same band occupation.
in all the layers and the influence of temperature on the interlayer exchange coupling 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.

V. DISCUSSION OF RESULTS

The calculated interlayer exchange coupling (IEC) using the modified RKKY method at two different band occupations (n=0.2, 0.8) are presented in Figs. 2 and 3. The IEC oscillates between ferro- and antiferromagnetic configurations. This behaviour is in agreement with that of the trend seen in many experimental works. 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.

![Fig. 1 Geometry of the M/N/M multilayer with a spacer thickness of 5 monolayers](image)

![Fig. 2 The interlayer exchange coupling (IEC) obtained from the modified RKKY method at various values of the spacer thickness at a band occupation of 0.2 (T=0K)](image)

![Fig. 3 The interlayer exchange coupling (IEC) obtained from the modified RKKY method at various values of the spacer thickness at a band occupation of 0.8 (T=0K)](image)

Our interest in this study is to point out 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 Figs. 4 and 5. 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 (Fig.4) whereas it is ferromagnetic for the correlated film (Fig.2). The behaviour 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.

Fig.4 The interlayer exchange coupling (IEC) obtained from the conventional RKKY method at various values of the spacer thickness at a band occupation of 0.2 (T=0K)

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 as well as the conventional RKKY methods. The results are plotted in Fig.6. 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 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 Fig.6. The modified RKKY method further predicts that the IEC gets saturated after 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 of 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 Fig.6 is not obtainable from his method. The IEC is found to be sensitive to the band occupation also and it may be seen from the Figs. 2, 3 and 4, 5. 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 Figs.2-5 are the Fourier fit 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 as 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 Figs.2-5 that there is a overall sign change of the decaying wave from the modified RKKY to conventional RKKY 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 compared to that of the conventional RKKY.

VI. CONCLUSIONS

We have evaluated the interlayer exchange coupling 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 ferro- 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 sophisticated than the conventional RKKY method and hence a calculation for real local moment multilayers can be expected give reliable and interesting results.

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