ORNL/CP-9602

CONF-980102--

First Principles Calculations of Interlayer Exchange Coupling in bcc Fe/Cu/Fe Structures.

M. Kowalewski, B. Heinrich
Simon Fraser University, Burnaby, B.C. V5A-1S6 Canada

T. C. Schulthess, W. H. Butler
Oak Ridge National Laboratory, Oak Ridge, Tennessee
37831-6114 USA

Abstract — We report on theoretical calculations of interlayer exchange coupling between two Fe layers separated by a modified Cu spacer. These calculations were motivated by experimental investigations of similar structures by the SFU group. The multilayer structures of interest have the general form: Fe/Cu(k)/Fe and Fe/Cu(m)/X(1)/Cu(n)/Fe where X indicates one AL (atomic layer) of foreign atoms X (Cr, Ag or Fe) and k, m, n represent the number of atomic layers of Cu. The purpose of the experimental and theoretical work was to determine the effect of modifying the pure Cu spacer by replacing the central Cu atomic layer with the atomic layer of foreign atoms X. The first principles calculation were performed using the Layer Korringa-Kohn-Rostoker (LKKR) method. The theoretical thickness dependence of the exchange coupling between two semi-infinite Fe layers was calculated for pure Cu spacer thicknesses in the range of 0<k<16. The effect of the foreign atoms X on the exchange coupling was investigated using the structure with 9 AL Cu spacer as a reference sample. The calculated changes in the exchange coupling are in qualitative agreement with experiment.

Index Terms — ferromagnetic and antiferromagnetic interlayer exchange coupling, magnetic moment, The Layer Korringa-Kohn-Rostoker method, ultrathin magnetic multilayers.

I. METHOD OF CALCULATION

The first-principles electronic calculations were performed within the local spin density approximation to the density functional theory using the Layer Korringa-Kohn-Rostoker (LKKR) technique [1]. The LKKR technique requires only two dimensional periodicity and is therefore ideally suited for the self-consistent treatment of infinite sandwich systems. These trilayer systems were constructed by embedding the varying number of bcc Cu (001) spacer atomic layers between two semi-infinite bcc Fe (001) crystals. The heterogeneous Fe/Cu(modified)/Fe structures consisted of 9 Atomic Layer (AL) of copper with the central Cu replaced by one AL of Fe, Cr or Ag. Across each entire sandwich the lattice structure of bcc Fe was used with the exception of one calculation for silver where the Cu lattice was allowed to relax to accommodate the larger Ag atoms. All the interfaces were taken to be atomically abrupt and smooth. We have used the atomic sphere approximation for the potentials which were self-consistently relaxed in the spacer atomic layers as well as in 4 iron atomic layers at the interfaces. All other Fe layers were fixed to the bulk potentials of bulk bcc iron. In all self-consistent calculations we have used s, p and d partial waves, 16 energy points on a semi circular contour in the complex energy plane, 36 special k-points in 1/8th section of the 2D Brillouin zone and 37 plane waves for the interlayer scattering.

We used the frozen potential approximation to calculate the exchange coupling in the following way. The same interface potentials are used to calculate the band energies of the ferro and antiferromagnetic configurations, respectively. Using Andersen's Force Theorem [2], the interlayer exchange coupling, J, is then calculated from the difference between these band energies, E_{band}, plus an additional term that consists of the product of the charge difference between the two states, dq, and the electro-chemical potential, μ:

$$J=E_{band(AF)} - E_{band(FM)} + dq^*\mu \qquad (1)$$

The number of points in the k-space integral to calculate the band energies was greatly increased and details of the convergence will be discussed in the next section.

II HOMOGENEOUS SPACER TRILAYERS: bcc Fe/Cu/Fe(001)

The convergence of our calculations for each thickness of the homogeneous copper spacer is illustrated in Fig. 1, where the coupling as a function of the spacer thickness is plotted for 36, 136, 528 an 2080 special k-points in the Irreducible Brillouin Zone (IBZ). It is clear that for 2080 special k-points the solution is convergent. Our results are in full agreement with the earlier theoretical studies of bcc Cu(001) performed by Herman et al. [3] whose studies were, however, limited to thicknesses between 5-9 AL. Our calculations of coupling between 1-15 AL allowed us to compare the theoretical models for the oscillatory exchange coupling by Edwards et al. [4], Bruno [5], Stiles [6] and Slonczewski [7]. In Fig. 2 we plotted the interlayer exchange coupling as a function of the copper spacer thickness and fitted the data with two cosine functions of different amplitudes and a common $1/d^2$ thickness dependence:

$$[A_1\cos[(2\pi d/\lambda_1)-\phi_{10}] + A_2\cos[(2\pi d/\lambda_2)-\phi_{20}]]/d^2 \qquad (2)$$

where d is the thickness of the spacer in atomic layers (AL), A_i, ϕ_{i0}, and λ_i are the amplitude, phase and period of oscillations (in AL), respectively, for the short (i=1) and long (i=2) range oscillations. We used spacer thicknesses of d>5 where the above asymptotic formula is likely to be accurate [4]. The short and long periods of oscillations of $\lambda_1=2.1\pm 0.2$

Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corporation.

DISCLAIMER

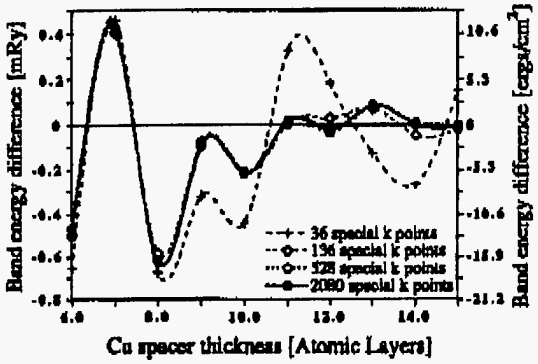

Fig. 1. The band energy difference between ferro and antiferromagnetic configurations plotted as a function of the Cu spacer thickness for increasing number of special k-points in the 2D irreducible Brillouin zone. All lines are cubic spline fits to guide the reader's eye. The calculations are convergent for 2080 special k-points.

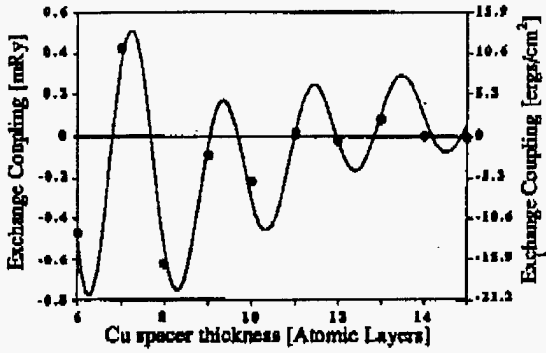

Fig. 2. Interlayer exchange coupling plotted as a function of Cu spacer thickness for atomically smooth interfaces. Our theoretically calculated points are fitted with the sum of two cosine functions of different amplitude and a common $1/d^2$ thickness dependence to determine the short and long period oscillations.

AL and λ_2=10.0 ±1.0 AL in our fit are in good agreement with those predicted by Bruno for pure bcc Cu(001) spacer using extremal spanning vectors along Γ-X, Γ and M lines in the Fermi surface, see [5].

To simulate the possible variation of the thickness of the spacer due to the interface roughness we ~~simulated the interfacial roughness by~~ averaging the coupling over several adjacent spacer thicknesses. It is reasonable to take the final thickness of the copper spacer to contain approximately 50% of the nominal thickness with some of the spacer being thinner and some thicker [8,9,10]. Therefore, we calculated the moving average of the earlier calculated points for the roughness confined to three topmost layers, see Fig. 3. Comparison of the calculated results in Fig. 3 with experimental points in Fig. 21 of [10] shows striking similarities in the behavior of the coupling. However, there are also notable differences. The cross-over from ferromagnetic to antiferromagnetic coupling occurs at ~6.5 AL in theoretical calculation and ~8.5 AL in experimental data. Moreover, the maximum antiferromagnetic coupling in theoretical calculation occurs at ~9 AL whereas in

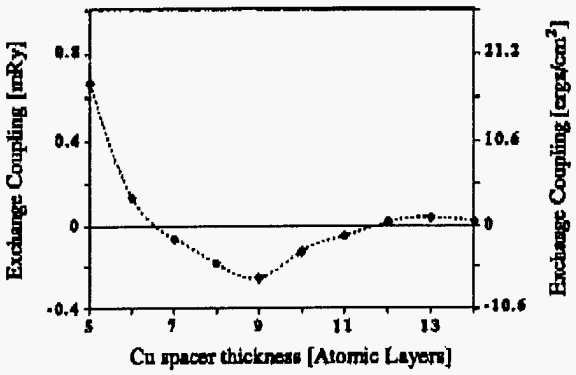

Fig. 3. The interlayer exchange coupling as a function of the Cu spacer thickness for rough interfaces. The exchange coupling values in this graph were obtained by calculating a moving average of the couplings for pure Cu spacer with ideally smooth interfaces. The interfacial roughness was simulated by averaging over values of the exchange coupling, J(N), for three adjacent pure Cu spacer thicknesses: J_N=0.25J(N-1)+0.5J(N)+0.25J(N+1), where J_N is the interlayer exchange coupling across the Cu spacer with simulated rough interfaces. The line through the points is a cubic spline fit to guide the reader's eye. Notice that the short period oscillations are suppressed.

experiment at ~11 AL. The two atomic layer shift in both features between the theory and the experiment can be attributed to the interface roughness and suggests that up to 2 AL of Cu are effectively removed from the pure Cu spacer. This is not surprising: the Cu atomic layers at the interface which contain Fe atoms ~~are~~ most likely ~~excluded from the coupling~~. The equivalent of 1 AL of Cu at each interface is removed from participation in the coupling across the spacer due to the interface roughness. The experimental structures with nominal spacer thickness N lose two atomic layers resulting in the effective ~~coupling across~~ N-2 atomic layers of the spacer. Further Mossbauer ~~experimental~~ studies to investigate this possibility are underway and will be reported elsewhere.

as magnetic layers rather than spacer layers.

III. HETEROGENEOUS SPACERS: bcc Fe/Cu(4)X(1)Cu(4)/Fe(001)

The structure with pure 9 AL copper spacer was chosen as the reference sample in this set of calculations. Atomically abrupt interfaces were used at both Fe/Cu and Cu/Fe interfaces as well as at the interfaces of the single foreign atomic layer X with the surrounding copper matrix to simplify the calculations. Foreign atoms of Fe, Cr and Ag were used. Table I lists all the theoretical values of the exchange coupling and magnetic moments of the relevant foreign atoms in the central spacer. The value of the exchange coupling for the pure 9 AL copper spacer is listed as the first entry in Table I for comparison. First, Fe, Cr and Ag were placed in the unrelaxed Cu lattice and the exchange couplings calculated, see entries 2,3 and 4 respectively in Table I. For Fe and Cr atoms each volume per atom in the lattice structures (and Wigner-Seitz radius) is nearly the same as that for Cu. However, the lattice constant of Ag is about 10% larger, and, therefore, in order to accommodate Ag atoms the Cu lattice was relaxed. Silver atoms were assumed

TABLE I
SUMMARY OF RESULTS FOR HETEROGENEOUS Cu SPACER
The interlayer exchange coupling and the magnetic moment per foreign atom for the heterogeneous spacer where the central atomic Cu layer was replaced by the atomic layer of foreign atoms of Fe, Cr and Ag. The average magnetic moment per atom in the fully disordered Fe is zero.

FOREIGN ATOM	Exchange Coupling in mRy (ergs/cm^2)	Magnetic Moment in Bohr magneton
1. pure Cu	-0.090 (-4.8)	-0.004
2. Fully Ordered Fe	+0.60 (+32)	+2.55
3. Cr	+0.24 (+13)	+0.25
4. Unrelaxed Ag	-0.086 (-4.6)	-0.002
5. Relaxed Ag	+0.036 (+1.9)	-0.002
6. Fully Disordered Fe	-0.040 (-2.1)	±2.55

to maintain the in-plane lattice spacing of bcc Cu(001) and only the lattice spacing between the two Cu atomic layers next to silver was expanded vertically to accommodate for the correct volume per silver atom. The result of this calculation is shown as entry 5 in Table I. The final sixth entry in Table I is the calculation of exchange coupling for one atomic layer of Fe with fully disordered moments. The magnetic disorder was modeled by a disordered alloy which consisted of equal amounts of Fe that have moments pointing in the + and - z directions, respectively. The disorder was treated within the coherent potential approximation (CPA) [11].

The results listed in Table I show that the foreign Fe atomic layer in the ordered magnetic state with the strongest average moment per atom affects the coupling the most, the magnetically ordered Cr atomic layer has an intermediate effect with magnetic moment per atom an order of magnitude smaller and Ag has the least effect with approximately zero magnetic moment per atom. The experimental Mossbauer studies of Fe/Cu(5)/Fe(1)/Cu(5)/Fe sample indicated that the full atomic layer of Fe within the copper spacer is partially magnetically ordered [9]. Furthermore, similar studies of a Fe/Cu(5)/Fe(0.5)Cu(0.5)/Cu(5)/Fe sample showed that the atomic layer with 50% of Fe and 50% of Cu at the center of the copper spacer is magnetically disordered, see [9]. The first sample had a strong ferromagnetic coupling whereas the second sample maintained antiferromagnetic coupling, similar to the homogeneous Cu spacer, however, at approximately half its value. The theoretical calculation for the fully ordered and disordered Fe foreign layer, entry 2 and 6 in Table I, confirm that the magnetic ordering of foreign atoms is the most significant factor affecting the sign and strength of the exchange coupling. The fully ordered Fe layer changes the coupling to ferromagnetic similarly to the partially ordered Fe atomic layer in the experiment. Additionally, the magnetically disordered model calculation for an iron atomic layer at the center of the spacer yields an antiferromagnetic coupling which is approximately half the value of the coupling across the pure Cu spacer, just as in the experiment. These results indicate that the interlayer exchange coupling depends very strongly on the magnetic state of the atoms within the spacer. Obviously atoms within the spacer possessing some degree of magnetic ordering scatter the majority and minority valence electrons within the copper spacer in a different manner affecting not only the value but also the sign of the exchange coupling. Following Stiles and Bruno theory of interlayer exchange coupling, the spin asymmetry of the reflection coefficients for majority and minority spins in the quantum well of the spacer is the major variable in determining the sign and the magnitude of the coupling across the homogeneous spacers (see equation (13) in [6] and equation (5.27) in [5]). It is not surprising that placing magnetically ordered layers or atoms within the homogeneous spacer introduces further potential barriers for spins up and down of the valence electrons thus affecting the spin asymmetry within the spacer that may even change the sign of the coupling.

A less significant but nevertheless visible effect on the coupling is the change of the width of the quantum well of the spacer when the lattice relaxations are included in the case of Ag, see entries 4 and 5 in Table I. In the case of the unrelaxed Cu spacer lattice (entry 4 in Table I) the exchange coupling did not affect the coupling. This result is not surprising since the valence band of Ag is similar to that of Cu. However, when the Cu spacer is relaxed (entry 5 in Table I) the coupling changes to weakly ferromagnetic. The mechanism by which the coupling is affected is now related to changes in the width of the quantum well that the Cu valence electrons are traversing.

The experimental results did not show a crossover to FM coupling for either Cr or for Ag foreign atoms deposited in the center of the Cu spacer. We attribute these differences between the theoretical and experimental results to the fact that Fe, Cr and Ag atoms are not coherently placed in a single central atomic layer in the experimental structures, but are rather alloyed over a few central Cu layers.

REFERENCES

[1] J. M. MacLaren, S. Crampin, D. D. Vvedensky, J. B. Pendry, "Layer Koringa-Kohn-Rostoker technique for surface and interface electronic properties", Phys. Rev. B 40, p. 12164 (1989).
[2] (a) A. R. Machintosh, O. K. Andersen, " Electrons at the Fermi surface", ed. M. Springford , Cambridge Univ. Press, London, 1980, p. 149.
(b) M. Methfessel, J. Kubler, " Bond analysis of heats of formation: application to some group VIII and IB hydrides", J. Phys. F: Met Phys. 12, p. 141 (1982).
(c) Magnetic version of Forced Theorem: A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, V. A. Gubanov, "Local spin density functional approach to the theory of exchange interactions in ferromagnetic metals and alloys", J. Magn. Magn. Mat. 67, p. 65 (1987).
[3] F. Herman, J. Sticht, M. Van Schilfegaarde, " Long and short oscillatory exchange coupling in Fe/Cu and Co/Cu magnetic Multilayers", Mater. Res. Symp. Proc. 231, (1992).
[4] D. M. Edwards, J. Mathon, R. B. Muniz, M. S. Phan, "Oscillations of the exchange coupling in magnetic multilayers as an analog of de Haas-van Alphen effect", Phys. Rev. Lett. 67, pp. 493-496 (1991).
[5] P. Bruno, "Theory of interlayer magnetic coupling", Phys. Rev. B 52, pp. 411-439 (1995).
[6] M. D. Stiles, "Exchange coupling in magnetic heterostructures", Phys. Rev. B 48, pp. 7238-7258 (1993).
[7] J. C. Slonczewski, "Overview of interlayer exchange theory", J. Magn. Magn. Mat. 150, pp. 13-24 (1995).
[8] M. Kowalewski, B. Heinrich, K. Totland, J. F. Cochran, S. Govorkov, D. Atlan, K. Myrtle, P. Schurer ; "Studies of exchange coupling in Fe/Cu/Fe(001) 'loose spin' structures", Mater. Res. Soc. Symp. Proc. 384, p. 171 (1995).
[9] M. Kowalewski, B. Heinrich, J. F. Cochran; " Studies of interlayer exchange coupling in Fe/Cu/Fe ultrathin heterostructures", J. Appl. Phys. 81, p. 3904 (1997).
[10] B. Heinrich, J. F. Cochran, "Ultrathin metallic magnetic films: magnetic anisotropies and exchange interactions", Advances in Physics 42, pp. 523-639 (1993).
[11] J. B. Staunton, B. L. Gyorffy, D. D. Johnson, F. J. Pinski and G. M. Stocks, in "Alloy Phase Stability", G. M. Stocks and A. Gonis eds., Kluwer, Dordrecht, p. 469 (1989).

Report Number (14) ORNL/CP-96024
CONF-980102--

Publ. Date (11) 199801
Sponsor Code (18) DOE/ER, XF
UC Category (19) UC-400, DOE/ER

DOE