Calculation of electronic and magnetic structures of Fe/Cr, Co/Cr, and Ni/Cr multilayers

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We have performed realistic tight-binding band calculations for a series of $(X)_3/(\mathrm{Cr})_m/(X)_3$ sandwiches with bcc [001] interfaces, for X=Fe, Co, and Ni and m=3, 4, and 5. The moment distributions on the X and Cr layers have been determined self-consistently. In all the cases investigated, we have obtained locally stable, multiple solutions in which the ferromagnetic and antiferromagnetic couplings are realized between magnetic moments on successive X layers across intervening Cr layers. The similarities and differences in the magnetic properties among the three sandwiches are discussed. We also discuss implications of the results of our calculations on the magnetoresistance data recently obtained for Fe/Cr and Co/Cr multilayers.

I. INTRODUCTION

Magnetic multilayers consisting of transition metals and/or rare-earth metals have attracted continuous scientific interest in the last few years. One of the current topics in this field is the occurrence of a giant magnetoresistance effect. Baibich *et al.*[1] found that the reistivity of Fe/Cr multilayers is reduced by 46% when an external field of 20 kOe is applied. Various experiments, such as light scattering,[2,3] spin-polarized low-energy diffraction (SPLEED),[4] neutron diffraction,[5] and magnetization measurement,[3,6,7] show that the total magnetic moments on successive Fe layers couple antiferromagnetically across intervening Cr layers when the Cr-layer thickness is less than about 20 Å.[6,7] Recently Parkin *et al.*[8] reported oscillations in Fe-Fe couplings with a period of 18–20 Å out to a Cr-layer thickness of 50 Å. This antiferromagnetic coupling between Fe layers is expected to play an essential role in the large magnetoresistance observed.[9–11] When an external field is absent or small, the coupling between successive Fe layers is antiferromagnetic and the spin-dependent scattering at interfaces leads to large resistivity. Under a strong applied field, on the other hand, Fe moments order ferromagnetically and the resistivity is reduced because of a decrease in spin-dependent scattering. This antiferromagnetic Fe-Fe coupling cannot, however, be understood within the localized (Heisenberg or Ising) model. If the localized model is applied to the Fe/Cr multilayer, the coupling between the Fe layers is ferromagnetic (FM) or antiferromagnetic (AFM), depending on whether the number of Cr layers is odd or even.[12] This is in contradiction with experimental results showing AFM Fe-Fe coupling.[6–8]

Quite recently, Levy *et al.*[10] and Hasegawa[13] performed band calculations for Fe/Cr multilayers. It was pointed out that the antiferromagnetic coupling is conceivable even when the number of Cr layer, m, is *odd*. In this case, the energy cost due to the unfavorable moment distributions in Cr layers is compensated by a change in the magnitude of the Cr moments;[10,13] the itinerant-electron system has an extra degree of freedom varying the magnitude of the magnetic moments whereas it is fixed in the localized model. Hasegawa[13] calculated also the interlayer exchange interaction, J, between Fe layers, which is positive for the nearest-neighbor layers as expected, but negative for $1 \leq m \leq 6$. This result is consistent with the observed Cr-thickness dependence of J.[6–8]

Since discovery of magnetoresistance in Fe/Cr multilayers, similar phenomena have been found in many transition-metal multilayers, such as Co/Cr,[8] Co/Ru,[8] Co/Au,[14] and Co/Cu/Ni-Fe/Cu.[15] Among them, we pay special attention to the Co/Cr multilayer, which is isomorphic with the Fe/Cr multilayer already discussed. It was reported that an applied field reduces the resistivity of Co/Cr multilayers by about 2.5%,[8] which is much smaller than the 46% reported for Fe/Cr multilayers.[1] Now several questions arise. Why is the magnetoresistance in Fe/Cr multilayers much more significant than that in Co/Cr multilayers? Is the magnetoresistance in Co/Cr multilayers expected to arise from the AFM coupling between Co moments as is the case in Fe/Cr multilayers, although no experimental evidence has yet been reported? It is therefore interesting to make a theoretical study on the similarities and differences between Fe/Cr and Co/Cr multilayers, which is the purpose of the present paper. We will report calculations of the electronic and magnetic structures in a series of $(X)_3/(\mathrm{Cr})_m/(X)_3$ sandwiches, with X=Fe, Co, and Ni and m=3, 4, and 5.

The outline of this paper is as follows. In Sec. II we briefly describe the model and computational method which are used in the present study and which have been employed in our previous calculations.[13,16–18] The calculated moment distributions are reported in Sec. III. Section IV is devoted to supplementary discussions. Our conclusions are summarized in Sec. V.

II. MODEL AND COMPUTATIONAL METHOD

We assumed a sandwich in which both X and Cr atoms lie on a common bcc lattice with [001] interface. The configuration of the multilayer under consideration is

given by[19]

$$(Cu)_2/(Cr)_2/(X)_3/(Cr)_m/(X)_3/(Cr)_2/(Cu)_2/Cu(001)\ ,$$

with the semi-infinite Cu(001) simulating the nonmagnetic substrate employed in the experiments. The layer parallel to the (001) interface is assigned by the index n, which is assigned the value 1 for the top layer. Crystalline anisotropy was neglected for computational simplicity. For a given multilayer, we employed the model Hamiltonian

$$H = H_0 + H_I\ , \tag{1}$$

where H_0 and H_I denote the one-electron and interaction terms. The one-electron part is expressed by the tight-binding d-band Hamiltonian given by

$$H_0 = \sum_{\sigma}\sum_{j}\sum_{m} E_j a^{\dagger}_{jm\sigma} a_{jm\sigma} + \sum_{\sigma}\sum_{j,j'}\sum_{m,m'} t^{mm'}_{jj'} a^{\dagger}_{jm\sigma} a_{j'm'\sigma}\ , \tag{2}$$

where $a^{\dagger}_{jm\sigma}$ ($a_{jm\sigma}$) is a creation (annihilation) operator of a σ-spin electron of the orbital m on the lattice site j, E_j is the core potential, and $t^{mm'}_{jj'}$ stand for the two-center transfer integrals which are given, after canonical band theory,[20] as

$$\left.\begin{matrix}(dd\sigma)\\(dd\pi)\\(dd\delta)\end{matrix}\right\} = \left.\begin{matrix}-6\\4\\-1\end{matrix}\right\} \times (W_d/2.5)(S/R)^5\ . \tag{3}$$

Here R is the interatomic distance, S the Wigner-Seitz radius, and W_d is the d-band-width parameter. Transfer integrals are included up to second-nearest-neighbor sites, and those between different kinds of atoms are assumed to be given by their geometrical averages.[20]

The interaction term in Eq. (1) is given by

$$H_I = \tfrac{1}{4}\sum_{j}(U_j N_j^2 - J_j M_j^2)\ , \tag{4}$$

where N_j (M_j) denotes the charge (magnetic-moment) operator on site j, and U_j and J_j are Coulomb and exchange interactions, respectively. We assumed that $U_j = J_j$ so as to reduce the number of parameters and treated them within the Hartree-Fock approximation. The U values for Fe and Co were chosen such that we obtain the ferromagnetic ground state with magnetic moments of $2.2\mu_B$ and $1.7\mu_B$, respectively. We chose the U value for Cr so as to obtain the commensurate antiferromagnetic states with the sublattice moment of $0.6\mu_B$. These are consistent with the local-spin-density-functional calculations for bcc Fe,[21] Co,[22] and Cr.[23] The U values for Ni and Cu were assumed to be the same as those of Fe. The number of d electrons were taken as 5.0, 7.4, 8.4, 9.4, and 10.0 per atom for Cr, Fe, Co, Ni, and Cu, respectively. The core potentials E_j were chosen to preserve local charge neutrality, with neglect of slight deviations near the interfaces. The band parameters (W_d, U_n, and E_n) employed in our calculations are summarized in Table I.[24]

TABLE I. Parameters used in the calculations.

Atom	E_n (eV)	W_d (eV)	U_n (eV)
Cr	5.80	8.00	0.67
Fe	3.80	6.12	0.80
Co	2.78	5.44	0.80
Ni	1.70	4.76	0.80
Cu	0.00	4.08	0.80

We assumed that all atoms on the nth layer have the same averages of local magnetic moments and number of electrons, M_n and N_n, which were calculated self-consistently by an iterative method.[13,16–18] The iteration was continued until the initial and resultant M_n and N_n agree within an assumed respective accuracy of $0.025\mu_B$/atom and electrons/atom. By adopting a variety of initial trial solutions, we repeated our calculations, looking for solutions which are locally stable in configuration space.

III. CALCULATED RESULTS

A. Fe/Cr/Fe sandwiches

First we discuss results for Fe/Cr/Fe sandwiches. We obtained the FM and AFM solutions, both of which are numerically stable. Figures 1(a) and 1(b) show, respec-

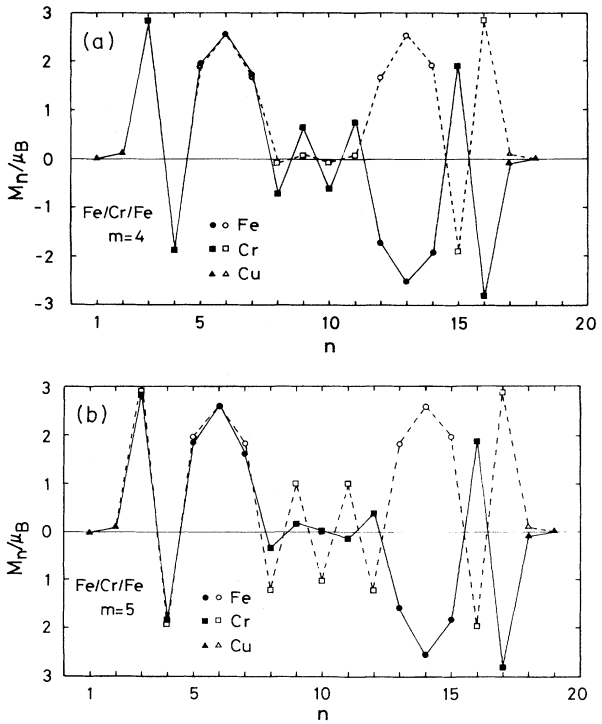

FIG. 1. Local moments in the AFM (solid curve) and FM (dashed curve) solutions in $(Fe)_3/(Cr)_m/(Fe)_3$ sandwiches with (a) $m=4$ and (b) $m=5$.

tively, the calculated moment distributions in $(Fe)_3/(Cr)_m/(Fe)_3$ sandwiches with $m=4$ and 5. The moment distributions are almost symmetric or antisymmetric with respect to the center of the sandwiches. The substrate Cu layers have no local moments while interfacial Cu layers are slightly spin polarized because of the molecular field arising from adjacent Fe layers. The calculated moment distributions on Fe and Cr layers have the following features.[19]

(i) In both AFM and FM solutions, the Fe regions are ferromagnetic while the Cr regions are antiferromagnetic.

(ii) Magnetic moments on the central and interfacial Fe layers are $2.5\mu_B$ and $1.5\mu_B$ respectively, both in the FM and AFM solutions.

(iii) Magnitudes of Cr moments in the FM solutions for $m=3$ and 5 and in the AFM solutions for $m=4$ are about $1.0\mu_B$. In these cases, the spin-density wave (SDW) on Cr layers is compatible with the moment distribution on Fe layers. In the other cases where the Cr SDW is incompatible with Fe moments, the Cr moments become very small.

(iv) The moment distribution of the AFM solution for odd m ($=3$ and 5) has a node at the central Cr layer.

(v) The coupling between interfacial Fe and Cr moments is antiparallel both in the AFM and FM solutions.

These features in the present calculation are the same as those obtained in a previous study for Fe/Cr/Fe sandwiches,[13] for which we adopted the configurations given by

$$(Cu)_3/(Fe)_3/(Cr)_m/(Fe)_3/(Cu)_3/Cu(001)\ ,$$

slightly different from those used in the present study. Cr layers have been subsequently added outside Fe/Cr/Fe sandwiches as buffers, by which Fe moments on the outside interfaces (at $n=5$ and $n=m+10$) in the new calculation is reduced ($2.0\mu_B$) compared with that ($2.5\mu_B$) in the previous one.[13] Our results based on the tight-binding model are similar to those obtained with first-principles calculations for a $(Fe)_3/(Cr)_5$ superlattice.[10] The enhanced moments on Fe and Cr layers are attributed to the two-dimensional character of the sandwiches.[26]

B. Co/Cr/Co sandwiches

Next we discuss the calculated result of Co/Cr/Co sandwiches, for which we again obtained locally stable AFM and FM solutions. The calculated moment distributions in $(Co)_3/(Cr)_m/(Co)_3$ sandwiches with $m=4$ and 5 are shown in Figs. 2(a) and 2(b), respectively. We note the following features.

(i) Magnitudes of Co moments are $(1.0-1.5)\mu_B$, which is reduced relative to the bulk value of $1.7\mu_B$.

(ii) When the Cr SDW is compatible with Co moments in the FM solutions for $m=3$ and 5 and in the AFM solutions for $m=4$, the magnitude of Cr moments is $(0.2-0.5)\mu_B$.

(iii) The moment distribution of the AFM solutions for odd m ($=3$ and 5) has a node at the central Cr layer.

(iv) The coupling between the interfacial Co and Cr moments is rather complicated. The coupling is parallel in the AFM solutions for $m=3$ and 5 while it is antiparallel in the FM solution for $m=4$. In the AFM solution

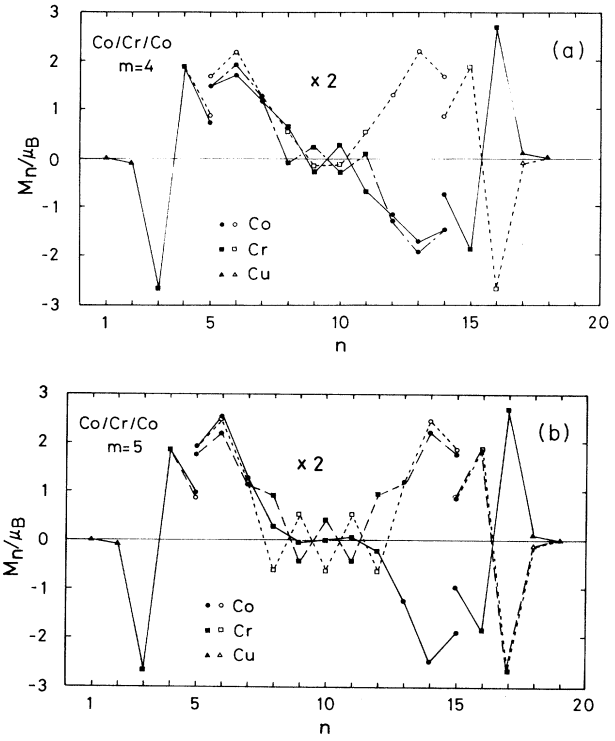

FIG. 2. Local moments in the AFM (solid and chain curves) and FM (dotted and dashed curves) solutions in $(Co)_3/(Cr)_m/(Co)_3$ sandwiches with (a) $m=4$ and (b) $m=5$. Note that the scale of magnetic moments in the inner Co/Cr/Co layers has been multiplied by a factor 2.

for $m=4$ and in the FM solution for $m=5$, we obtained not only parallel but also antiparallel alignments in interfacial Co and Cr moments.

Thus, we obtained much more stable solutions in Co/Cr/Co sandwiches than in Fe/Cr/Fe sandwiches. First-principles calculations for a $(Co)_3/(Cr)_5$ superlattice[26] yield the FM solution similar to ours for the corresponding sandwich.

C. Ni/Cr/Ni sandwiches

The calculated moment distributions in $(Ni)_3/(Cr)_m/(Ni)_3$ sandwiches with $m=4$ and 5 are plotted in Figs. 3(a) and 3(b), respectively. They have the following features.

(i) Although the Cr regions are antiferromagnetic, the Ni regions are not ferromagnetic, having peculiar moment distributions.

(ii) Magnitudes of Ni moments are less than $0.3\mu_B$, which is smaller than the bulk value of $0.6\mu_B$.

(iii) Magnetic moments on Cr layers in the AFM solutions for $m=3$ and 5 and in the FM solution for $m=4$ are $(1.2-1.8)\mu_B$, which are much enhanced compared with the bulk value. In the other cases, the magnitudes of Cr moments are less than $0.2\mu_B$.

(iv) The coupling of the interfacial Ni and Cr moments

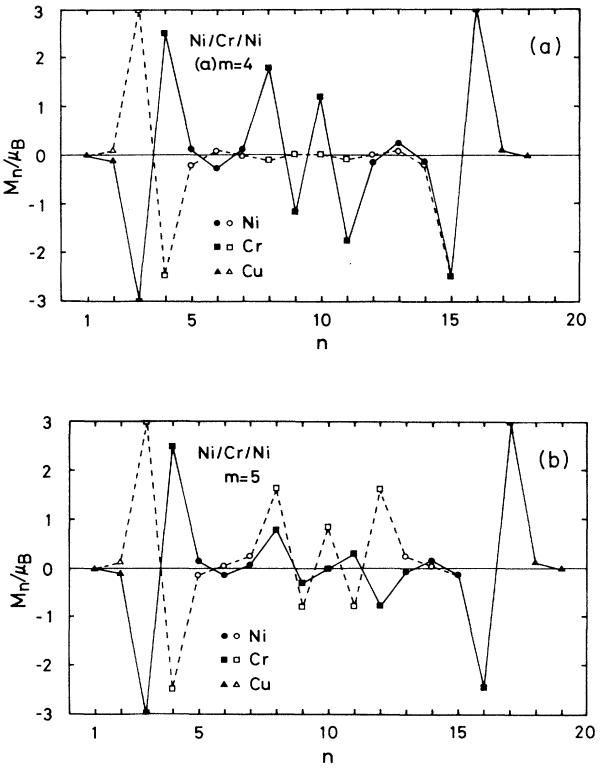

FIG. 3. Local moments in the AFM (solid curve) and FM (dashed curve) solutions in $(Ni)_3/(Cr)_m/(Ni)_3$ sandwiches with (a) $m=4$ and (b) $m=5$.

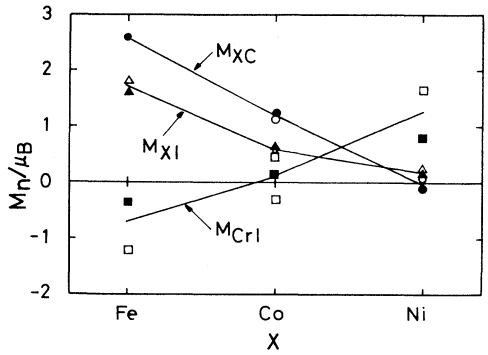

FIG. 4. Magnetic moments on the central X layers (M_{XC}), those on the interfacial X layers (M_{XI}), and those on the interfacial Cr layers (M_{CrI}), in $(X)_3/(Cr)_5/(X)_3$ sandwiches against X (=Fe, Co, and Ni). The filled (open) marks denote the results of the AFM (FM) solutions. The curves are only a guide for the eye.

is parallel in all the cases.[27]

In Ni/Cr/Ni sandwiches Ni moments are nearly quenched because of the influence of the Cr layers, though Cr moments can be enhanced compared with its bulk value, as noted in item (iii) above. Magnitudes of magnetic moments on outer Cr layers sandwiched by Cu and Ni, are ($2.5-3.0\mu_B$), which is more enhanced than those on inner Cr layers.

IV. DISCUSSIONS

It is interesting to investigate the similarities and differences in the magnetic properties among the $X/Cr/X$ sandwiches, with X=Fe, Co, and Ni. We plot, in Fig. 4, the X dependence of magnetic moments at the central X layer (M_{XC}), those at the interfacial X layer (M_{XI}), and those at the interfacial Cr layer (M_{CrI}). This shows the trend in magnetic properties of $X/Cr/X$ sandwiches with changing X. Magnitudes of magnetic moments on the central and interfacial X layers decrease as going from X=Fe to X=Ni. We note that the interfacial Cr moments are antiparallel to M_{XI} for X=Fe while they are parallel for X=Ni.[27] In the case of X=Co we obtained two solutions corresponding to parallel and antiparallel alignment of interfacial Co and Cr moments, as discussed in Sec. III B. Figure 4 clearly shows that magnetic properties are rather sensitive to the choice of X atom in the three sandwiches.

In order to establish which of the multiple solutions we have obtained is most stable, it is necessary to make a comparison of their ground-state energies. We have tried it, but could not determine the most stable solutions because the energy difference is too small compared with our computational accuracy. Note that the energy difference is expected to be of the order of the saturation field, H_s, whose typical value is 10 kOe ($\sim 10^{-3}$ eV),[1] while the total d-band energy is of the order 10 eV. In order to by-pass this difficulty, we calculated in Ref. 13, the interlayer exchange interaction, $J_{nn'}$, between the nth and n'th layers in Fe/Cr superlattices as a function of m, the number of intervening Cr layers. The exchange interaction $J_{nn'}$ is positive for the nearest-neighbor pair ($m=0$) as expected. It is nearly vanishing for $m=1$, and becomes negative for $2 \leq m \leq 6$, suggesting that the AFM solutions become more stable than the FM solutions. Thus the antiferromagnetic Fe-Fe coupling observed in Fe/Cr/Fe multilayers with Cr thickness less than 20 Å (Refs. 6–8) can be understood in terms of the exchange interactions between d electrons. In order to explain the Ruderman-Kittel-Kasuya-Yosida (RKKY)-type long-range oscillation in J observed in thicker Cr layers,[8] it would be necessary to take into account s-electron and s-d hybridization,[28,29] which are neglected in our calculation of Ref. 13.

It was reported that the magnetoresistance is large (46% reduction) in Fe/Cr multilayers[1] while it is fairly small (2.5% reduction) in Co/Cr multilayers.[8] Our calculation might explain the difference in the magnetoresistance between Fe/Cr and Co/Cr multilayers as follows. According to existing theories,[9–11] the magnetoresistance arises from the spin-dependent scatterings at the interfaces. The difference in the exchange potentials at the interface between X and Cr layers, V, given by

$$V=\tfrac{1}{2}|U_X M_{XI}-U_{Cr}M_{CrI}| \cong (U/2)\delta M \ , \qquad (5)$$

provides a measure of the spin-dependent interfacial scattering potential, where $U=U_X \cong U_{Cr}$ and

$\delta M = |M_{XI} - M_{CrI}|$. Our calculation shows that in Co/Cr/Co sandwiches, we get $\delta M = (0.1-0.8)\mu_B$ because $M_{CoI} = (0.5-0.6)\mu_B$ and $M_{CrI} = -(0.2-0.5)\mu_B$. On the other hand, in Fe/Cr/Fe sandwiches, $M_{FeI} = (1.6-1.8)\mu_B$ and $M_{CrI} = -(0.4-1.2)\mu_B$ lead to $\delta M = (2.0-2.8)\mu_B$, which is much larger than that in Co/Cr/Co sandwiches. Since the magnetoresistance is more significant when V is larger,[9-11] it is probable that the difference in δM *qualitatively* explains the difference in the observed magnetoresistance between Fe/Cr and Co/Cr multilayers.

V. CONCLUSIONS

We have calculated the electronic and magnetic structures of $(X)_3/(\mathrm{Cr})_m/(X)_3$ sandwiches with X=Fe, Co, and Ni and m=3, 4, and 5. In all the cases investigated we have obtained the AFM and FM solutions, both of which are locally stable in the configuration space. It has been shown that the AFM solutions can be stable even when the number of intervening Cr layers is *odd* within the itinerant-electron model.[10,13] This is realized by the characteristics of the itinerant-electron model with an extra degree of freedom changing the magnitude of local moments. Cr layers are expected to play important roles for the sandwiches to have multiple solutions.[30] On the basis of our calculations, we have qualitatively discussed the observed difference in magnetoresistance in Fe/Cr and Co/Cr multilayers. Our calculations have shown that an applied field to the multilayers with an *odd* number of Cr layers induces the SDW in the Cr layers. Observation of the *field-induced SDW* by experiments, such as neutron diffraction, will be interesting. Further experimental studies on magnetic properties and magnetoresistance are required not only for Fe/Cr multilayers but also for Co/Cr and Ni/Cr multilayers.

ACKNOWLEDGMENTS

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