Interlayer magnetic coupling: Effect of alloying in the spacer

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The influence of alloying in the spacer on the periods and the amplitudes of the oscillations of exchange coupling in magnetic multilayers is studied from first principles. The effect of substitutional randomness in the spacer is treated within the coherent potential approximation and the validity of the simplified virtual-crystal approximation is examined. As a case study, results obtained for Co(001) slabs in random fcc $Cu_{1-x}M_x$ spacers (M=Ni, Zn, and Au) are presented and discussed. [S0163-1829(96)52630-5]

The interlayer exchange coupling (IEC) between magnetic layers separated by a nonmagnetic nonrandom spacer has recently been the subject of intense theoretical studies, particularly on the *ab initio* level.[1,2] The study of the effect of alloying in the spacer represents another powerful experimental and theoretical tool[3–6] to verify existing models[7] of the IEC by continuously varying the medium between the magnetic layers. Conventional band structure methods are of limited use for such studies although in very particular cases an application of the virtual-crystal approximation (VCA) may be justified, as e.g., for VCr and CrMn alloy spacers studied recently.[8] In general–as to be expected–the complete neglect of alloy disorder makes a reliable determination of the coupling amplitudes and, to some extent, also of the coupling periods, uncertain even in such favorable cases. The Green's function formulation of the IEC, however, is capable to treat randomness within the coherent potential approximation (CPA) which, in turn, is known to describe reliably compositional trends in random alloys. In the present paper the Lloyd formulation of the IEC as based on a spin-polarized surface Green's function (SGF) technique in terms of the tight-binding linear muffin-tin orbital method is used (for further details, see Ref. 2).

Our model consists of two magnetic subsystems (left and right, denoted by $\mathcal{L}$ and $\mathcal{R}$) each containing M magnetic layers on the top of a semi-infinite nonmagnetic spacer. The spacer slab of varying thickness N, which separates magnetic layers, is considered as a part of the subsystem $\mathcal{R}$. Nonmagnetic spacer layers are formed by a binary alloy $A_{1-x}B_x$ of composition x. If ϑ denotes a particular relative angle between spin directions in the magnetic subsystems $\mathcal{L}$ and $\mathcal{R}$, the exchange energy $\mathcal{E}_x$ is defined as $\mathcal{E}_x=\overline{\Omega}(\pi)-\overline{\Omega}(0)$, where the configurational averaged grand canonical potentials $\overline{\Omega}(\vartheta)$ are given by[9]

$$\overline{\Omega}(\vartheta)=\frac{1}{\pi N_\parallel}\mathrm{Im}\sum_{\mathbf{k}_\parallel}\int_C f(z)\,\mathrm{trln}\left(1-\frac{1-\cos(\vartheta)}{2}M(\mathbf{k}_\parallel,z)\right)dz. \tag{1}$$

Here $f(z)$ is the Fermi-Dirac distribution function and tr denotes the trace over angular momentum indices. In Eq. (1), the energy integration is performed over a contour in the upper half of the complex energy plane, the sum runs over $\mathbf{k}_\parallel$ vectors in the irreducible surface Brillouin zone, and $N_\parallel$ is the corresponding unit surface area. The quantity $M(\mathbf{k}_\parallel,z)$ is defined as[10]

$$\begin{aligned}M=&-(1-S_{10}\overline{\mathcal{G}_{\mathcal{L}}^{\uparrow}}S_{01}\overline{\mathcal{G}_{\mathcal{R}}^{\uparrow}})^{-1}S_{10}(\overline{\mathcal{G}_{\mathcal{L}}^{\uparrow}}-\overline{\mathcal{G}_{\mathcal{L}}^{\downarrow}})\\&\times(1-S_{01}\overline{\mathcal{G}_{\mathcal{R}}^{\downarrow}}S_{10}\overline{\mathcal{G}_{\mathcal{L}}^{\downarrow}})^{-1}S_{01}(\overline{\mathcal{G}_{\mathcal{R}}^{\uparrow}}-\overline{\mathcal{G}_{\mathcal{R}}^{\downarrow}}),\end{aligned} \tag{2}$$

where matters of simplicity the arguments $\mathbf{k}_\parallel$ and z have been omitted. In Eq. (2) the quantities $S_{01}(\mathbf{k}_\parallel)$ and $S_{10}(\mathbf{k}_\parallel)$ are the structure constants which couple neighboring layers, $\overline{\mathcal{G}_{\mathcal{S}}^{\sigma}}(\mathbf{k}_\parallel,z)$, $\mathcal{S}=\mathcal{L},\mathcal{R}$, is the configurationally averaged SGF of the magnetic subsystem and σ denotes the spin index ($\sigma=\uparrow,\downarrow$). It should be noted that in the presence of randomness the expression for the IEC is formally analogous to the case of a nonrandom spacer in the sense that the SGF's which enter Eq. (2) are substituted by the corresponding configurational averages.[11] The theoretical basis for this simplification—which reduces computational times by almost two orders of magnitude—relies on the ''alloy force theorem''[12] and on the ''vertex cancellation theorem.''[13]

In here, numerical studies are presented for an fcc Cu spacer as alloyed with Ni or Zn or Au. This choice is motivated by the fact that alloying with Ni or Zn alters the electron concentration and, consequently, modifies the topology of the alloy Fermi surface, which in the asymptotic limit, is closely related to the coupling periods in terms of Fermi

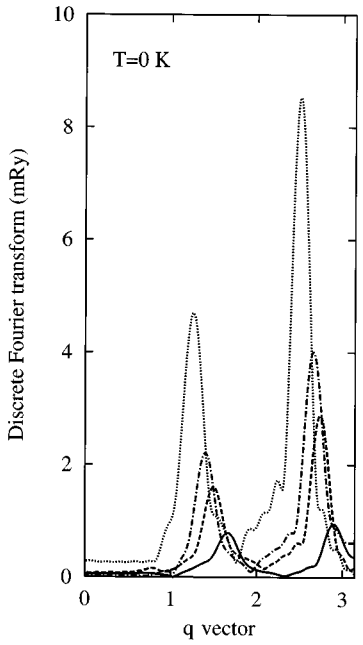

FIG. 1. Composition dependence of the absolute values of a discrete Fourier transform of $\mathcal{E}_x$ at $T=0$ K for two Co(001) slabs each five monolayers thick separated by an fcc $Cu_{1-x}Ni_x$ alloy spacer: (i) $Cu_{0.75}Ni_{0.25}$ (solid line), (ii) $Cu_{0.85}Ni_{0.15}$ (dashed line), $Cu_{0.9}Ni_{0.1}$ (dashed-dotted line), and (iv) an ideal Cu spacer (dotted line).

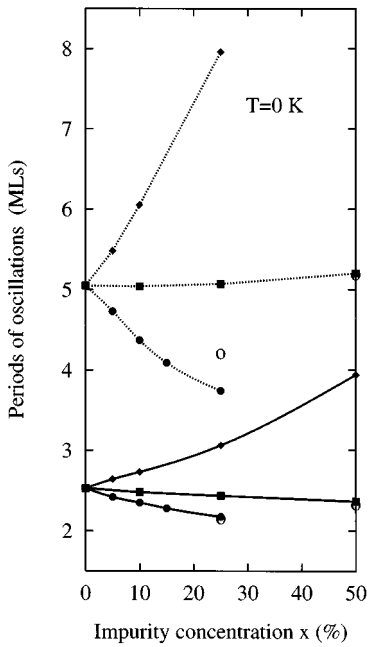

FIG. 2. Composition dependence of the coupling periods at $T=0$ k for two Co(001) slabs each five monolayers (ML's) thick separated by an fcc $Cu_{1-x}M_x$ alloy spacer: (i) $M=$Ni (bullets), (ii) $M=$Au (squares), and (iii) $M=$Zn (diamonds). The lines serves as a guide for the eye and distinguish between short (solid lines) and long (dotted lines) period oscillations. Open circles for $Cu_{0.75}Ni_{0.25}$ and $Cu_{0.5}Au_{0.5}$ represent the approximate virtual-crystal values. The periods are given in ML's.

surface spanning vectors.[14] The changes in the coupling periods can then be viewed as a contraction (Ni) or the expansion (Zn) of the alloy Fermi surface. On the contrary, alloying of Cu with Au does not alter the electron concentration. In all cases, however, the coupling amplitudes are expected to be influenced by the presence of alloy disorder. Strictly speaking, in random alloys the Fermi surface is not well defined. In particular cases, however, when the alloy Fermi energy lies in the sp part of the spectrum above the d-band complex, alloy broadening usually is weak causing therefore only little smearing of the Fermi surface. As it is obvious from Eqs. (1) and (2), the present Green's function formulation does not rely upon the existence of the alloy Fermi surface.

As magnetic systems, two Co(001) slabs each 5 monolayers (ML) thick and, in particular cases, two semi-infinite Co(001) slabs sandwiching the alloy spacer, were studied. The alloy lattice constants were estimated from the experimental values for bulk constituents assuming Vegard's law. No layer relaxation at interfaces between magnetic and spacer layers is considered, i.e., we assume an ideal fcc lattice in the whole space. All calculations were performed for $T=0$ K, numerical details of which can be found in Refs. 2 and 9.

A discrete Fourier transform $F(q)$ was performed for a set of $N^2\mathcal{E}_x(N)$ values, where N is the number of spacer layers. The periods of oscillations p_i (in monolayers) were identified from the positions q_i of pronounced peaks of $|F(q_i)|$ as $p_i=2\pi/q_i$. The amplitudes of oscillations A_i were estimated from $A_i=(2/n)|F(q_i)|$, where n is the number of $N^2\mathcal{E}_x(N)$ values used in the Fourier analysis (typically N varies from 10–50, i.e., $n=40$). This is illustrated in Fig. 1 for the case of 5 ML thick Co(001) slabs with an fcc $Cu_{1-x}Ni_x$ spacer. Alloying shifts corresponding peaks of $|F(q)|$ to higher values of q and suppresses their heights.

The results for the concentration dependence of the periods of the oscillations for the chosen alloy spacers are summarized in Fig. 2. For an ideal Cu spacer the short period oscillations (SPO) and the long period oscillations (LPO) are 2.53 MLs and 5.05 MLs, respectively. Alloying of Cu with Ni decreases the electron concentration in the alloy and leads to a contraction of the alloy Fermi surface. In terms of the Ruderman-Kittel-Kasuya-Yosida (RKKY) formulation[14] this leads to an expansion of spanning vectors corresponding to an fcc(001) layer orientation. Both the SPO and the LPO are reduced with increasing Ni concentration but this reduction is more pronounced for the LPO. The present results for the LPO agree qualitatively with the simplified calculations of Ref. 4 based on a RKKY formulation and approximating the spanning vectors by a linear interpolation between the bulk band structures of Cu and Ni. It is therefore interesting to compare the present results with those performed within the same computational scheme but employing the VCA instead of the CPA. The results for $Cu_{75}Ni_{25}$ are shown in Fig. 2 as open circles. As one can see, in particular for the LPO the shift of the coupling periods differs in the VCA from that in the CPA.

The opposite behavior, namely a shift to larger periods and again rather pronounced for the LPO, was found when alloying Cu with Zn, namely the case when the Fermi surface expands. Note that the periods of oscillations for a $Cu_{1-x}Zn_x$ spacer increase faster with x than they decrease in

TABLE I. Relative amplitudes $A(Cu_{1-x}M_x)/A(Cu)$ for two Co(001) slabs each five monolayers thick separated by an fcc $Cu_{1-x}M_x$ alloy spacer (M=Ni, Au, Zn). The values at T=0 K are given for both the short and the long periods.

x_{Ni}	short/long	x_{Au}	short/long	x_{Zn}	short/long
0.0	1.00/1.00	0.0	1.00/1.00	0.0	1.00/1.00
0.05	0.66/0.67	0.1	0.91/0.97	0.05	0.86/0.78
0.1	0.39/0.48	0.25	0.79/0.67	0.1	0.78/0.62
0.15	0.25/0.34	0.5	0.60/0.69	0.25	0.60/0.41
0.25	0.11/0.17			0.5	0.50/-

the case of $Cu_{1-x}Ni_x$. Finally, for a $Cu_{1-x}Au_x$ spacer even for large concentrations of Au (up to 50%), we observe a negligible concentration dependence of the periods of the oscillations which is consistent with the composition-independent average electron number in these alloys as well as with available experimental data.[5] From Fig. 2 it is obvious that the coupling periods obtained for a $Cu_{0.5}Au_{0.5}$ spacer the CPA and VCA are virtually the same.

The coupling amplitudes for random spacers relative to an ideal Cu spacer are given in Table I. The strong suppression of the coupling amplitudes in $Cu_{1-x}Ni_x$ spacers with increasing Ni content (about an order of magnitude for $Cu_{0.75}Ni_{0.25}$) can be viewed as increasing hybridization-induced disorder in the *sp* states since the Fermi energy moves towards the *d*-band complex. This is consistent with the deviations observed between the VCA and CPA for $Cu_{0.75}Ni_{0.25}$. For example, the coupling amplitudes for the SPO and LPO in $Cu_{0.75}Ni_{0.25}$ spacer as calculated within the VCA are 5.6 and 4.5 times larger than the corresponding CPA values, respectively. For the whole composition range in $Cu_{1-x}Au_x$ and $Cu_{1-x}Zn_x$ spacers the alloy Fermi level lies in the *sp* bands above the *d* bands of Cu, Au, and Zn. Consequently, for these alloy spacers disorder at the Fermi level is confined to the weak disorder in the *sp* states and is not influenced by the presence of strong *d*-band disorder in states well below the Fermi energy. The effect of disorder on the coupling amplitudes is thus weaker than in the case of $Cu_{1-x}Ni_x$ spacers. For example, the VCA amplitudes for the SPO and LPO for $Cu_{0.5}Au_{0.5}$ are only about 10% and 60% larger than the corresponding CPA values, respectively. Note, however, that for $Cu_{0.5}Zn_{0.5}$ the LPO are already nearly suppressed, or better, the corresponding peak in a discrete Fourier transform is comparable to the background noise. Adopting a RKKY-like picture, the SPO and the LPO periods are connected via so-called stationary points[14] located in different parts of the surface Brillouin zone. Quite likely, the different influence of alloying on the amplitudes of the SPO and the LPO is connected with the anisotropy of electron-impurity scattering at the alloy Fermi level. Such information can be obtained, e.g., from surface-state resonance experiments.[15] This particular aspect, however, requires a separate theoretical study.

The dependence of $\mathcal{E}_x$ on the spacer thickness N is illustrated in Fig. 3(a) for the case of two semi-infinite Co(001) slabs sandwiching $Cu_{0.75}Ni_{0.25}$, $Cu_{0.5}Zn_{0.5}$, and $Cu_{0.5}Au_{0.5}$ alloy spacers. In the limit of large thickness of Co slabs the LPO's are suppressed and the SPO's dominate.[2,16] An increase of the coupling period is clearly seen for

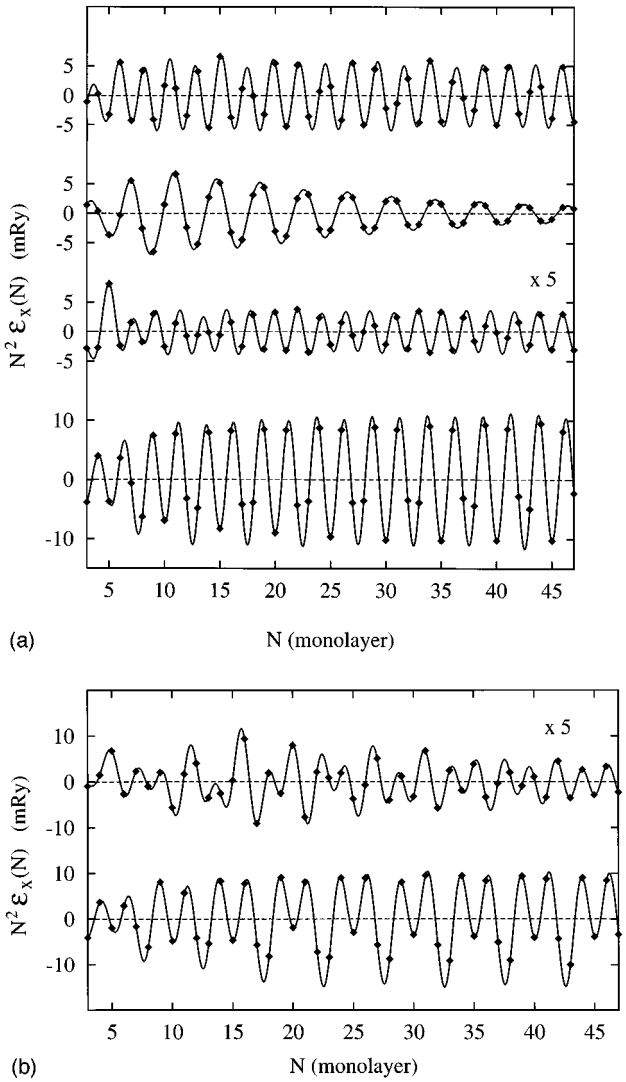

FIG. 3. Exchange coupling $N^2\mathcal{E}_x(N)$ at T=0 K as a function of the spacer thickness N: (a) Two semi-infinite Co(001) subsystems sandwiching a spacer of (from bottom to top) ideal Cu, $Cu_{0.75}Ni_{0.25}$ (multiplied by a factor 5), $Cu_{0.5}Zn_{0.5}$, and $Cu_{0.5}Au_{0.5}$; (b) two Co(001) slabs each five monolayers thick embedded into a spacer of (from bottom to top) ideal Cu and $Cu_{0.75}Ni_{0.25}$ (multiplied by a factor 5). Diamonds refer to the calculated values, the solid line (back Fourier transform) serves as a guide to the eye.

$Cu_{0.5}Zn_{0.5}$, while the reduction for a $Cu_{0.75}Ni_{0.25}$ spacer is less pronounced. The period of oscillations for a $Cu_{0.5}Au_{0.5}$ alloy spacer remains essentially unchanged. From Fig. 3(a) one can also see that the $Cu_{1-x}Ni_x$ and $Cu_{1-x}Au_x$ alloy spacers exhibit an RKKY-like behavior with the amplitudes being proportional to N^{-2}, however, with values reduced in comparison to the ideal Cu spacer. This reduction is particularly pronounced for a $Cu_{0.75}Ni_{0.25}$ spacer (compare Table I). An approximate RKKY-like behavior with a weak exponential damping $\approx\exp(-0.04N)$ was found for $Cu_{0.5}Zn_{0.5}$. Both the SPO and the LPO are present for the case of 5 MLs Co(001) slabs sandwiching a $Cu_{1-x}Ni_x$ spacer. The ratio between the SPO and the LPO periods, which is 1.85 for an ideal Cu spacer, is reduced to 1.2 for $Cu_{0.75}Ni_{0.25}$ because the period of the SPO is more reduced with respect to the Ni concentration than that for the

LPO. Consequently, due to ''incommensurate'' periods, the dependence of $\mathcal{E}_x$ on the spacer thickness N, Fig. 3(b), is now more complicated despite the fact that a discrete Fourier analysis gives again two well pronounced peaks corresponding to the LPO and the SPO (see Fig. 1).

In summary, we have performed a systematic *ab initio* study of the dependence of the coupling periods and amplitudes corresponding to $Cu_{1-x}M_x(001)$ spacers with $M=$Ni, Au, Zn by employing the vertex cancellation theorem. By varying the composition the electron concentration and, consequently, the Fermi surface can change which, in turn, gives rise to a shift for the coupling periods. Periods for $Cu_{1-x}Ni_x$ ($Cu_{1-x}Zn_x$) spacers are shifted to smaller (larger) values while for $Cu_{1-x}Au_x$ spacer they are essentially insensitive to the alloying. Alloy disorder leads to a suppression of the coupling amplitudes, particularly if the alloy Fermi energy moves towards the region of d bands like for $Cu_{1-x}Ni_x$ system. We also found that the $Co/Cu_{1-x}Au_x/Co(001)$ system can be described reasonably well within a simplified VCA approach while, for example, the $Co/Cu_{1-x}Ni_x/Co(001)$ system definitely requires a more accurate treatment in terms of the CPA.

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