

Exchange coupling between iron layers separated by silver and gold

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 (Received 23 May 1997; revised manuscript received 12 August 1997)

The exchange couplings between bcc Fe layers separated by fcc Ag and Au are calculated for Fe/Ag/Fe and Fe/Au/Fe (001) trilayer structures as functions of the spacer thickness for several temperatures. The calculated couplings show a short-period oscillatory behavior in Fe/Au for all temperatures investigated. For Fe/Ag a long period prevails for temperatures $T \geq 300$ K, but at $T=0$ a strong short-period contribution is present for Ag thicknesses ≥ 30 atomic planes. These results are in very good agreement with finite-temperature experiments, but the coupling strengths as calculated by assuming perfect interfaces are much larger than those observed. It is shown that interplanar distance relaxation at the interfaces leads to a rather large effective change of the coupling amplitude in Fe/Au for Au thicknesses ≤ 20 atomic planes, but mainly causes a phase shift in the oscillatory coupling for Fe/Ag. It is found that interfacial interdiffusion substantially reduces the amplitude of the coupling in Fe/Au/Fe, but not much in Fe/Ag/Fe. [S0163-1829(97)06645-9]

The lattice constant of bulk bcc Fe matches the nearest-neighbor distances of both fcc Ag and Au within less than 1%. This allows the growth of Fe/Ag and Fe/Au multilayers in the [001] direction with low-stress interfaces. In the stacking, the fcc Ag and Au (001) planes place themselves rotated by 45° around [001] relative to the Fe (001) planes. The distances between the fcc (001) planes in both Ag and Au ($d_{\text{Au(Ag)}}$) are $\approx \sqrt{2}$ times the bcc Fe (001) interplane spacing (d_{Fe}). The occurrence of atomic steps at the interfaces may thus cause significant misalignments. In fact, earlier attempts to measure the interlayer exchange coupling J in Fe/Ag/Fe multilayers failed to observe oscillatory dependence on Ag thickness,^{1,2} presumably due to the poor quality of the samples used. Later, however, J was measured in Fe/Ag/Fe and Fe/Au/Fe (001) structures with improved interfaces and, in both cases, it was found to oscillate, with decreasing amplitude, between ferro- and antiferromagnetic as a function of the spacer thickness N .^{3,4} Well-defined oscillations were also observed in these systems by scanning electron microscopy with polarization analysis.⁵ In those experiments, $J(N)$ predominantly oscillates with a long period in Fe/Ag/Fe (001), and with a short-period in Fe/Au/Fe (001).

For sufficiently large spacer thicknesses, the oscillation periods of $J(N)$ are related to the geometry of the spacer Fermi surface (FS).⁶⁻⁹ For fcc (001) noble-metal spacers such as Ag and Au, $J(N)$ has two oscillatory components: one with a long period coming from the "belly," and another with a short period associated with the "necks" of the spacer FS. The period of the former has been directly observed by photoemission in several noble-metal overlayers, including Ag and Au on bcc Fe (001).¹⁰ More recently, quantum well states around the FS necks were observed in Cu films grown on fcc Co (001).¹¹ Thus, it is currently also possible to probe the period of the neck contribution directly by photoemission.

The weight of each oscillatory component depends on the spacer FS curvatures and carrier velocities in the vicinity of the spacer FS extrema states.⁶⁻⁹ Comparison of the Fermi surfaces of Au and Ag, shows that at the belly the electron's average effective mass in Ag is ≈ 4 times larger than that in

Au; at the necks, however, they are comparable. On such a basis, one expects the long-period belly contribution to be much weaker in Au than in Ag. However, the relative importance of the two components in each system depends also on the degree of confinement experienced by carriers in those FS extremum states, caused by the magnetic layers.^{9,12-15} The (001) bcc/fcc interfaces involve two different lattice structures which are rotated by 45° around [001] relative to each other. Such a rotation imposes distinct boundary conditions on the spacer FS states at the interfaces, especially on those states around the necks, since the states at the belly are not affected by this rotation. A theoretical analysis of these effects requires explicit calculations of the coupling in these systems.

For perfect interfaces, the ions on every (001) atomic plane of the trilayer systems under consideration are arranged in a square lattice. Thus, the wave vector \vec{k}_{\parallel} parallel to the layers is a good quantum number. It follows that the formalism developed in Refs. 14 and 16 can be used to calculate J , defined as the total-energy difference per surface atom between the antiferromagnetic and ferromagnetic configurations of the trilayer. Most of the experimental results are for the bilinear exchange coupling term J_1 which for perfectly smooth Fe/Ag and Fe/Au (001) interfaces is virtually equal to $J/2$.¹⁶

To calculate J we have used a tight-binding model with s, p, d orbitals and hopping up to second nearest neighbors. The tight-binding parameters for all fcc Au and Ag planes were taken from Ref. 17, and those for ferromagnetic Fe were obtained as in Ref. 18. Results of $J_1(N)$ for Fe/Au/Fe and Fe/Ag/Fe (001) trilayers are shown in Fig. 1 for various temperatures. Clearly, $J_1(N)$ is dominated in Fe/Au by the short-period neck contribution, for all temperatures considered. This is evidenced in Fig. 2, where the discrete Fourier transform of $N^2 J$ is taken, at $T=0$ K, for large values of N . Such a procedure is useful for obtaining the relative amplitude of the various oscillatory components of J , provided one knows the asymptotic behavior of $J(N)$, and has a reliable method of calculating it in this region.¹⁹ The $1/N^2$

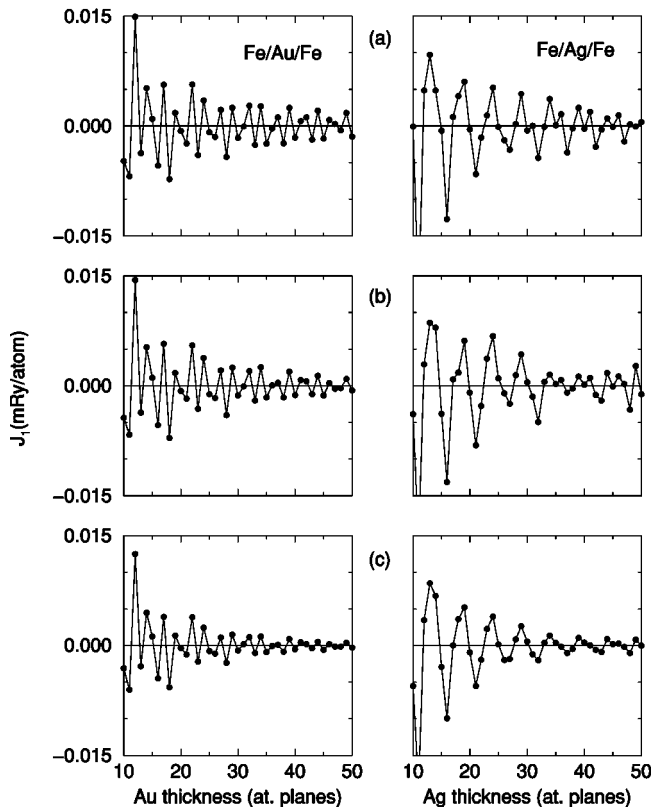


FIG. 1. Calculated exchange coupling for Fe/Au/Fe and Fe/Ag/Fe (001) trilayers as a function of spacer thickness for temperatures: $T=0$ K (a), $T=200$ K (b), and $T=400$ K (c).

asymptotic behavior of the coupling amplitude applies to $T=0$ K and ordered spacers only and, in most cases, it is not reached until $N>20$ atomic planes at least.

On the other hand, for Fe/Ag/Fe (001) trilayers it is the long-period belly contribution that clearly prevails for $T\geq 300$ K. At lower temperatures, however, a significant short-period contribution is visible in Fig. 1 for Ag thickness ≥ 30 atomic planes. The relative weight of both contributions is displayed in Fig. 2, where it is shown that at $T=0$ K, the short- and long-period components of $J(N)$ for Fe/Ag have comparable amplitudes asymptotically. The strong tempera-

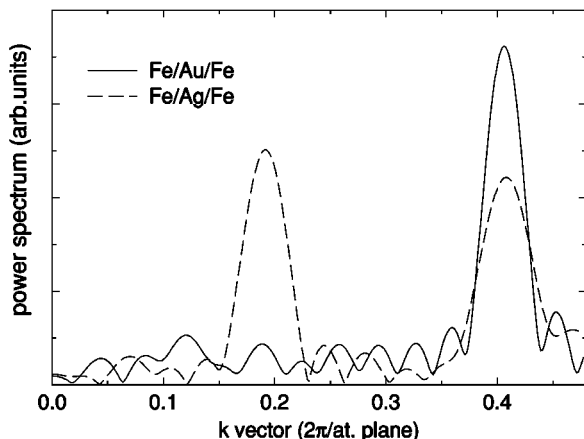


FIG. 2. Discrete Fourier transform (for $22\leq N\leq 50$) of $N^2 J(N)$ at $T=0$ K for Fe/Au/Fe (solid line) and Fe/Ag/Fe (dashed line) (001) trilayers.

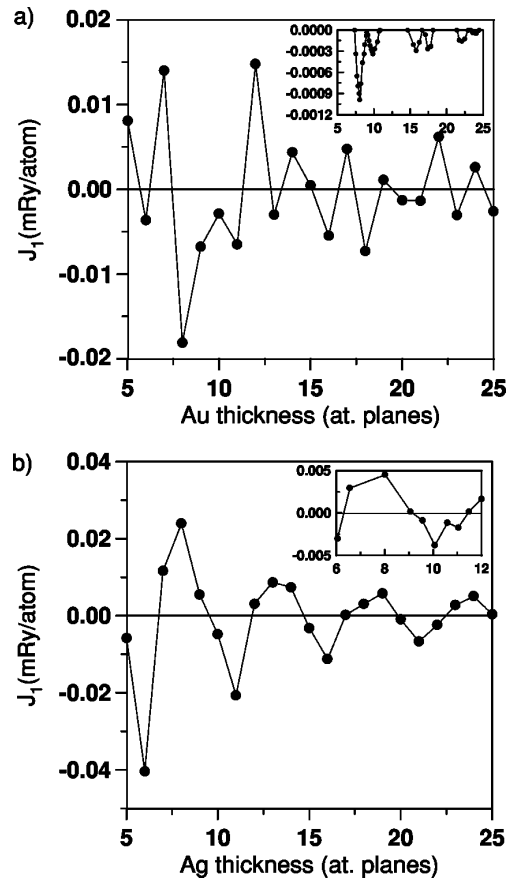


FIG. 3. Calculated exchange coupling at $T=300$ K for Fe/Au/Fe (a) and Fe/Ag/Fe (b) (001) trilayers as a function of spacer thickness. The insets show the experimental results of Fuss *et al.* (Ref. 4) (a), and of Celinski *et al.* (Ref. 3) (b).

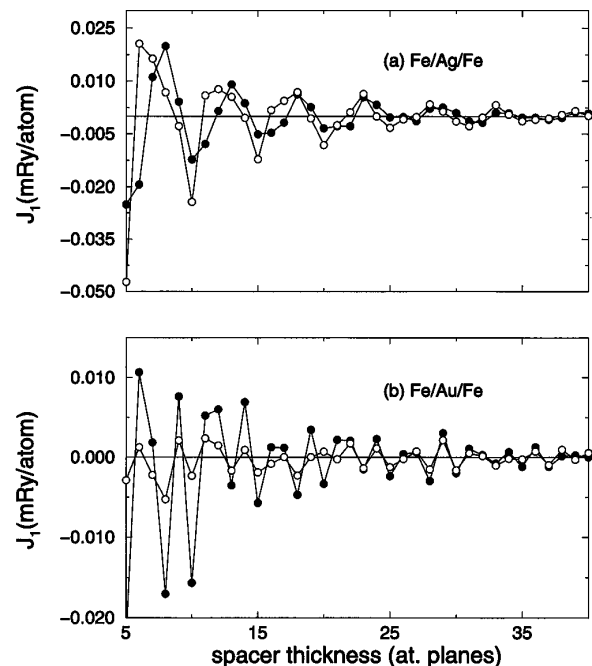


FIG. 4. Calculated exchange coupling for two different interfacial interplanar distances: $d_{\text{Fe-Sp}}=d_{\text{Fe}}$ open circles, and $d_{\text{Fe-Sp}}=d_{\text{Sp}}$ filled circles (see text). Results are obtained at $T=300$ K for Fe/Au/Fe (a), and Fe/Ag/Fe (b) trilayer systems, as a function of spacer thickness.

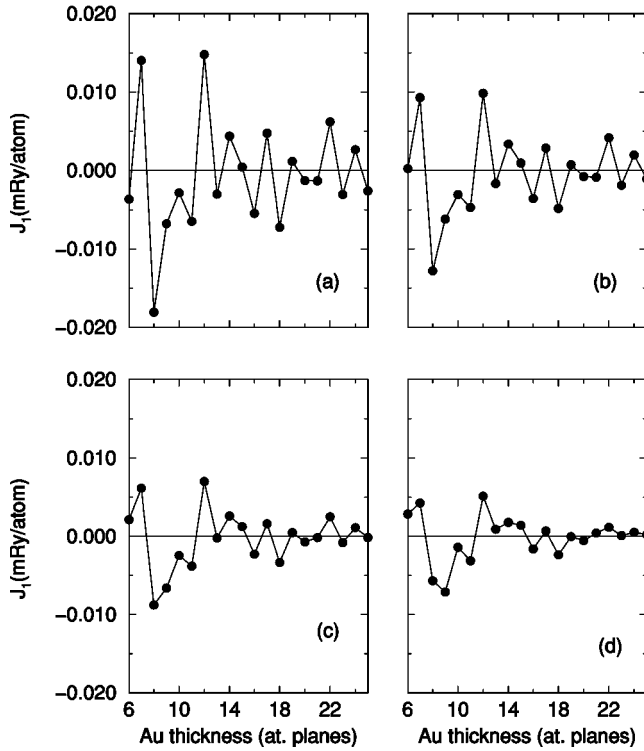


FIG. 5. Calculated exchange coupling for $\text{Fe}/\text{Au}/\text{Au}_{1-p}\text{Fe}_q/\text{Fe}_{1-q}\text{Fe}_q/\text{Fe}_{1-p}\text{Au}_q/\text{Fe}_{1-p}\text{Au}_q/\text{Fe}$ (001) trilayers. Results are obtained at $T=300$ K as a function of Au spacer thickness for $p=q=0$ (perfect interfaces) (a); $p=0.025$, $q=0.05$ (b); $p=0.05$, $q=0.1$ (c), and $p=0.1$, $q=0.15$ (d).

ture dependence of the short-period component, and the fact that it manifests itself at $T=0$ K for large values of N , suggest that the confinement mechanism discussed in Ref. 15 is relevant for the neck contribution in Fe/Ag trilayers. The periods determined from the position of the peaks in Fig. 2 agree perfectly with those calculated from the spacer FS extrema in the direction perpendicular to the layers, namely: $p_{\text{Au(Ag)}}^b = 9.2(5.3)$ atomic planes and $p_{\text{Au}}^n \approx p_{\text{Ag}}^n = 2.4$ atomic planes.

In Fig. 3 our results are compared with experiments. The agreement is excellent as far as the periods and phase of oscillations are concerned, but the calculated coupling strengths are much larger than those observed, both in Fe/Au/Fe and Fe/Ag/Fe (001) trilayers. The discrepancies may be due to interface roughness, which can drastically affect the coupling amplitude.²⁰ However, for Fe/Ag and Fe/Au (001), interplane distance relaxation near the interface can also play an important role, because of the relatively large difference between d_{Fe} and $d_{\text{Au(Ag)}}$. In fact, there is some evidence of tetragonal distortion of the Fe atoms at the interfaces in Fe/Ag superlattices.²¹ To investigate such an effect we have varied the interplanar distance $d_{\text{Fe-Sp}}$ between the Fe and spacer (Sp) (001) planes at the interface. We have considered two extreme cases, namely, $d_{\text{Fe-Sp}} = d_{\text{Fe}}$ and $d_{\text{Fe-Sp}} = d_{\text{Sp}}$. In each case, the tight-binding parameters have been scaled according to the distance-dependence prescription of Andersen *et al.*,²³ and the Fe-Sp hoppings were taken as the average between the Fe and spacer hoppings. The results are shown in Fig. 4, where one sees that interfacial interplane relaxation basically causes a phase shift, and a

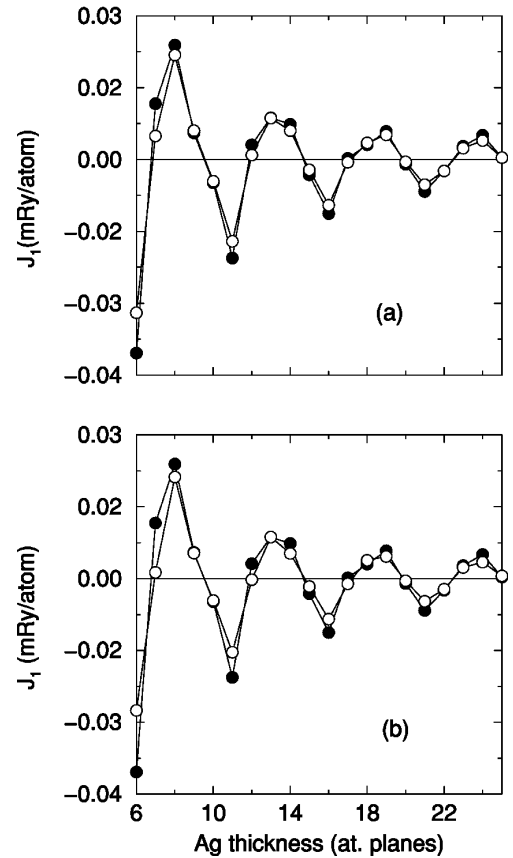


FIG. 6. Calculated exchange coupling for $\text{Fe}/\text{Ag}/\text{Ag}_{1-p}\text{Fe}_q/\text{Ag}_{1-q}\text{Fe}_q/\text{Fe}_{1-q}\text{Ag}_q/\text{Fe}_{1-p}\text{Ag}_q/\text{Fe}$ (001) trilayers. Results are obtained at $T=300$ K as a function of Ag spacer thickness for $p=0.025$, $q=0.05$ (a), and $p=0.05$, $q=0.1$. (b) Filled circles represent the results for perfect interfaces ($p=q=0$).

small change in the coupling amplitude of Fe/Ag/Fe (001), for small values of N . On the other hand, it has quite a large effect in Fe/Au/Fe, producing changes of a factor of ≈ 4 in the coupling strength for relatively small Au thicknesses. This apparently large variation in amplitude may also result, mainly, from a phase shift. Since the spacer is probed at discrete intervals, plane by plane, and J basically oscillates with a short period of about 2.4 atomic planes, a small phase shift, in this case, can effectively produce an apparent large change in amplitude.

Mossbauer spectroscopy has shown that some interdiffusion occurs, during the deposition of Fe over Ag and Au.²² The amount and extension of interfacial diffusion depend on the substrate's temperature during deposition. For growth at $T \leq 300$ K, it is expected to be limited and restricted to very few interfacial atomic planes, because Fe and Ag(Au) are known to be immiscible in the bulk. Actually, it differs from one interface to the other, as none seems to occur during the deposition of Ag on to Fe.^{21,22}

The occurrence of interdiffusion produces a disordered alloy at the interfaces. Translational symmetry parallel to the layers is then broken, and it is usually necessary to take configurational averages of the quantities of interest. As far as the interlayer coupling is concerned, Bruno *et al.*²⁴ have recently shown that, to an excellent approximation, one can still use Eq. (1) of Ref. 18 to calculate J across a disordered spacer, provided the Green functions involved are replaced

by their corresponding configurational averages. Kudrnovsky *et al.*²⁰ have discussed the effect of interface roughness in Co/Cu (001) systems. They found that the amplitude of the short-period component of the interlayer coupling is reduced, by nearly an order of magnitude, when only 10% of interfacial diffusion occur in Co/Cu/Co (001) trilayers. Here we investigate the effect of interfacial interdiffusion in a similar way, by treating the interfacial atomic planes as disordered alloys compatible with a given concentration profile. We restrict ourselves to small interfacial admixtures, and assume that it takes place at two planes on each side of the Au(Ag)/Fe interface only. The disorder is treated within a local average *t*-matrix approximation, which, in the dilute limit, is equivalent to the coherent-potential approximation used in Ref. 20. Our results for Fe/Au/Fe are presented in Fig. 5, for various interfacial alloy compositions. The reduction in the coupling amplitude, though large, is not as dramatic as that obtained in Co/Cu by Kudrnovsky, amounting to a factor of about two for 5–10 % Au/Fe interfacial interdiffusion. In Fig. 5 we see that for sufficiently large interface diffusion, a long-period oscillatory behavior begins to show up with the suppression of the short-period component, as expected. It is evident in Fig. 6 that for Fe/Ag/Fe, where the long-period component is dominant, the reduction in the coupling amplitude with the degree of interfacial diffusion is much less pronounced.

In summary, we have calculated the interlayer exchange coupling in Fe/Ag/Fe and Fe/Au/Fe (001) trilayers for several temperatures. We have found that the coupling oscillates with a long period in Fe/Ag, and with a short period in Fe/Au systems. Our results agree with experiment, as far as the periods of oscillations are concerned, but the coupling strengths as calculated by assuming perfect interfaces are much larger than those observed. We have shown that the effect of interplanar distance relaxation at the interfaces can be very important for relatively small spacer thicknesses. It leads to a rather large effective change of the oscillations amplitude in Fe/Au/Fe, but basically causes a phase shift in the oscillatory coupling for relatively small Ag thicknesses in Fe/Ag/Fe. We have also shown that a weak interfacial interdiffusion substantially reduces the coupling amplitude in Fe/Au/Fe, but has little effect in Fe/Ag/Fe. We conclude that a combination of interfacial interplane relaxation and interdiffusion, together with possible occurrence of terraces at the interfaces in real samples, probably accounts for the remaining discrepancies between calculated and observed values of the coupling strength in these systems.

We have benefited from helpful discussions with J. Mathon, D. M. Edwards, and M. V. Villeret. This work has been financially supported by CNPq and FINEP of Brazil.

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