Interlayer exchange coupling periodicity of various copper alloys in (111)-oriented FCC Co-CuX multilayers: Fermi surface effects

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Abstract. We report variations of between 10.7 and 15.8 Å in the periodicity of oscillatory interlayer exchange coupling of (111)-oriented $Cu_{1-x}Ge_x$–Co and $Cu_{1-x}Ni_x$–Co multilayers which depend on the germanium and nickel concentrations. These results agree with theoretical predictions that relate the exchange coupling periodicity to the diameter of the neck of the Fermi surface of Cu–Ge or Cu–Ni alloys.

1. Introduction

Oscillatory interlayer exchange coupling has been observed in several multilayer systems. These multilayers are based on 3d ferromagnetic layers (Fe, Co or Ni, or one of their alloys) separated by noble or transition-metal non-magnetic spacer layers. This oscillatory behaviour means that the zero-field configurations of the successive magnetic layers will be alternatively parallel (ferromagnetic (F) coupling) or antiparallel (antiferromagnetic (AF) coupling) when the thickness of the spacer layer is continuously increased.

One of the most studied system is the cobalt–copper system [1, 2]. It has very strong magnetoresistive properties: for antiferromagnetic coupled multilayers the giant magnetoresistance (GMR) exceeds 60% at room temperature.

The origin of the coupling in the case of noble-metal spacer layers has been addressed by Bruno and Chappert [4]. Their results can be summarized as follows: for a given crystallographic orientation of the spacer layer, the summation of the RKKY interactions over one plane of magnetic atoms in the ferromagnetic layers (plane perpendicular to the growth direction) induces selection rules; there is a finite number of wavevectors q_i which are allowed. These vectors are the nesting vectors that connect two points of the Fermi surface along the growth direction of the multilayer with opposite velocities. The asymptotic oscillatory expression for the interlayer coupling strength J_i versus the spacer thickness t is then

$$J_i(t) = \frac{J_0 \cos(q_i t + \phi)}{t^2} \tag{1}$$

where ϕ is the phase shift of the oscillations.

In fact, this theory is just the RKKY model but with a realistic nearly free-electron Fermi surface in the spacer layer and localized magnetic sites. The effect of the real band of

the transition metal and the effect of interface defects are totally neglected. This may be the reason why they could not correctly estimate the phase shift and the intensity J_0 of the coupling. However, their theory has a strong physical basis: the Fermi surface of the spacer layer. One of the main results of this theory is the correct prediction of a double-oscillation period for the (100) spacer orientation, as confirmed by Johnson *et al* [5] with (100)-oriented copper–cobalt samples. In the (111) orientation of the theory predicts only one coupling period (8.3 Å for pure copper) which is caused by a nesting vector that passes through the 'neck' of the Fermi surface along the (111) direction (figure 1).
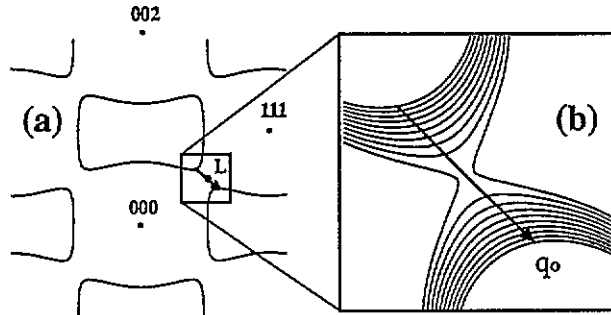

Figure 1. (*a*) Schematic representation of the (110) plane of the spacer layer in the reciprocal space (the nesting vector q_0 passing through the neck at L point is represented by an arrow). (*b*) Enlarged view of neck of the Fermi surface with several iso-energy curves ($\Delta E = 10$ mRyd) which correspond to lower values of E_F in the case of Cu–Ni alloys and to a reduced length of q_0.

Our group [6] and two other groups [7, 8] have investigated another way to test the theory of Bruno and Chappert with copper–cobalt-based multilayers. The idea is to replace copper ($Z = 29$) by an alloy with a suitable element (nickel ($Z = 28$)) which preserves its crystallographic structure and leads to minor modification of its band structure but which modifies its electronic density and therefore the shape of its Fermi surface.

The results that we present here concern (111)-textured Co–CuX multilayers prepared with two kinds of alloy: copper–nickel with up to 19 at.% Ni and copper–germanium ($Z = 32$) with up to 3.5 at.% Ge.

Copper–nickel alloys remain FCC solid solutions for any composition with a small lattice contraction towards nickel. They are non-magnetic up to 45 at.% Ni. On the contrary, germanium is only soluble into copper below 9.5 at.% where the alloys remain of FCC metal type. Our samples have been designed to stay inside these limits.

The simplest way (although certainly not exactly correct) to see what one expects in such alloys is to use the rigid-band model. In the hypothesis of one single conduction band, the effect of the nickel impurities in copper is to reduce the electronic density, and therefore the Fermi level. For a nickel atomic concentration x, the Fermi level is expected to be reduced to a first approximation as

$$E_F = \frac{\hbar^2}{2m}(3\pi^2 n_0)^{2/3}(1 - x)^{2/3} = E_{F_0}(1 - x)^{2/3}. \tag{2}$$

Germanium impurities are expected to play the opposite role, but with a three times larger increase in the electronic density.

Therefore, the shape of the Fermi surface will be modified in two opposite ways by Ni and Ge. In particular, the neck will become narrower in the case of nickel and wider in the case of germanium (figure 1). The consequence in real space is that we must have an increase in the coupling periodicity for $Cu_{1-x}Ni_x$–Co multilayer and a decrease for $Cu_{1-x}Ge_x$–Co multilayers. This is exactly the opposite behaviour of that predicted by the 'vernier' model which is based on the free-electron spherical Fermi surface.

2. Experimental procedures

As already published elsewhere [6,9], our samples are prepared at room temperature on Si (100) substrates with an Alcatel SCM 650 automated sputtering set-up. Our standard preparation conditions allow us to control the thickness of the layers with an accuracy and a reproducibility better than 1 Å. The roughness of the interfaces of our multilayers was estimated to be lower than 2 Å.

All the multilayers presented here have the nominal composition Si–Cr(50 Å)–[Co(11 Å)–Cu_{1-x}Ni(or Ge)$_x$(t Å)]$_{20}$–Cr(25 Å) where t is varied from 4 to 36 Å.

The deposition of the alloys was obtained from a composite target constituted of a copper disc of 100 mm diameter in which we have inserted various numbers of Ni or Ge rods of 5 mm diameter. The exact atomic composition was checked by quantitative microprobe analysis on alloys films 2000 Å thick with an accuracy better than 0.5%.

The alloys that we have obtained have the following concentrations: 81 at.% Cu–19 at.% Ni, 85 at.% Cu–15 at.% Ni, 90 at.% Cu–10 at.% Ni, 100 at.% Cu and 96.5 at.% Cu–3.5 at.% Ge. For this range of alloys, the electron-per-atom ratio varies from 0.81 to 1.11.

The thicknesses of the individual layers of our samples were measured after deposition by small-angle x-ray scattering (SAXS) which gave at least one Bragg peak corresponding to the multilayer stacking (figure 2). Large-angle scattering in $\theta/2\theta$ configuration essentially showed the (111) peak but with rather broad rocking curves. This effect should arise from some misorientation of our samples which are polycrystalline.
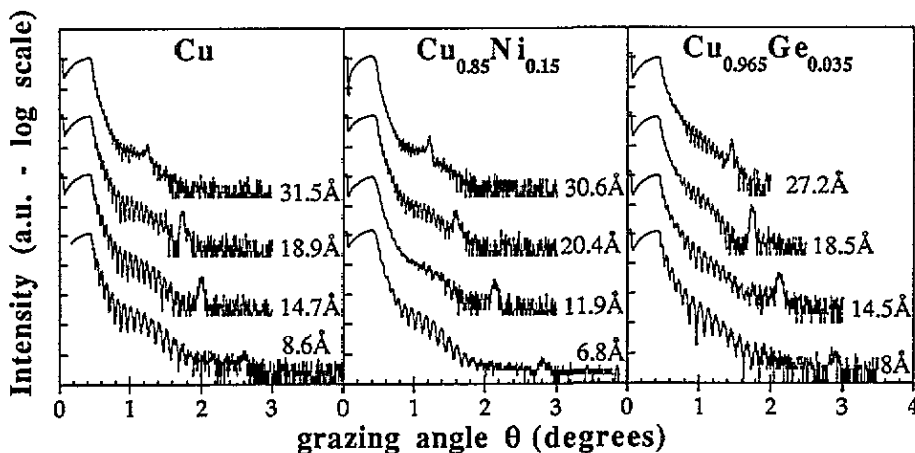
Figure 2. SAXS for several samples with the nominal composition Si–Cr(50 Å)–[Co(11 Å)–Cu_{1-x}Ni(or Ge)$_x$(t Å)]$_{20}$–Cr(25 Å) (a.u., arbitrary units). The three alloy compositions are indicated in the figure as well as the various spacer layer thicknesses.
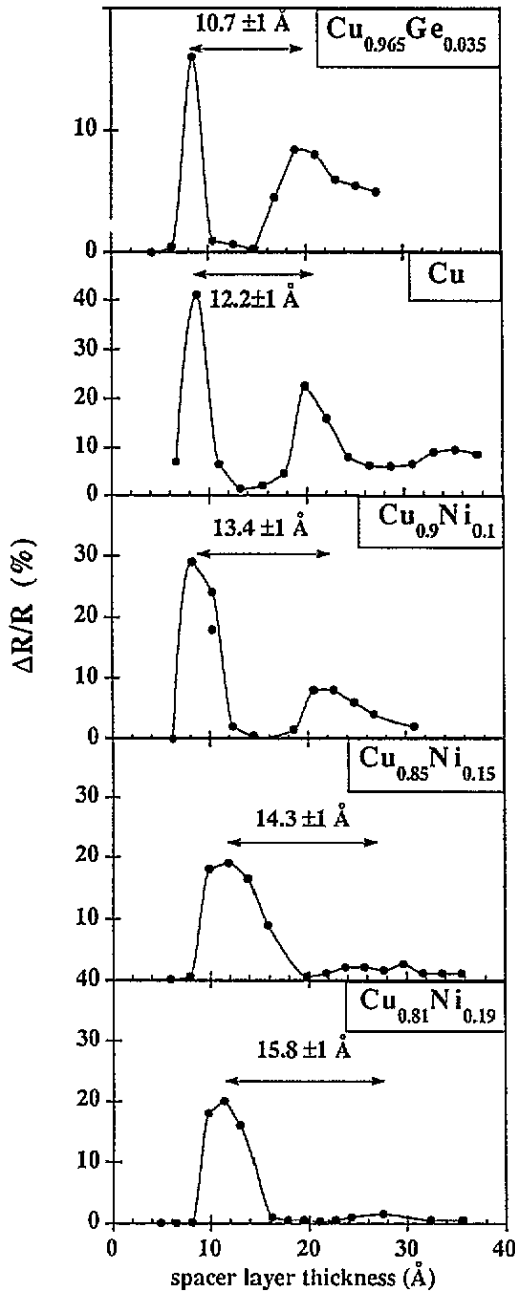
Figure 3. Room-temperature saturation magnetoresistance of our samples as a function of the spacer layer thickness.

The magnetoresistance was measured at room temperature with a rapid DC four-probe apparatus in fields up to 7000 Oe. Magnetization cycles were performed with a Quantum Design SQUID at 300 and 5 K up to 55 000 Oe.

3. Results and discussion

Our results concern the dependences of the interlayer coupling periodicity, of the AF coupling intensity, of the GMR and of the form of the magnetization curves on the Ni or Ge concentration.

3.1. Coupling periodicity

Figure 3 shows the saturation magnetoresistance at room temperature of our samples as a function of the spacer layer thickness. We clearly observe on the one hand a shift to larger spacer thicknesses of the two first GMR peaks for Cu–Ni alloys, and on the other hand a shift to lower spacer thicknesses for the Cu–Ge alloy.

One can define the interlayer coupling periodicity $\lambda_i = 2\pi/q_i$ as the distance between the two peaks. Following the theory of Bruno and Chappert, the vector q_i responsible for these oscillations makes an angle of 19° with the diameter of the Fermi surface neck. Its length is therefore about 6% larger. We have reported in figure 4 the deduced values of the neck diameter $q_N = 0.95q_i$ for our alloys. Our data are in fairly good agreement with rigid-band [10, 11] and KKR CPA [12] theoretical calculations and positron annihilation experiments [13–15]. It should be noted that our error bars which are due to the width of the GMR peaks and the thickness errors (±1 Å for periods ranging from 10 to 16 Å) are smaller than the positron annihilation error bars.
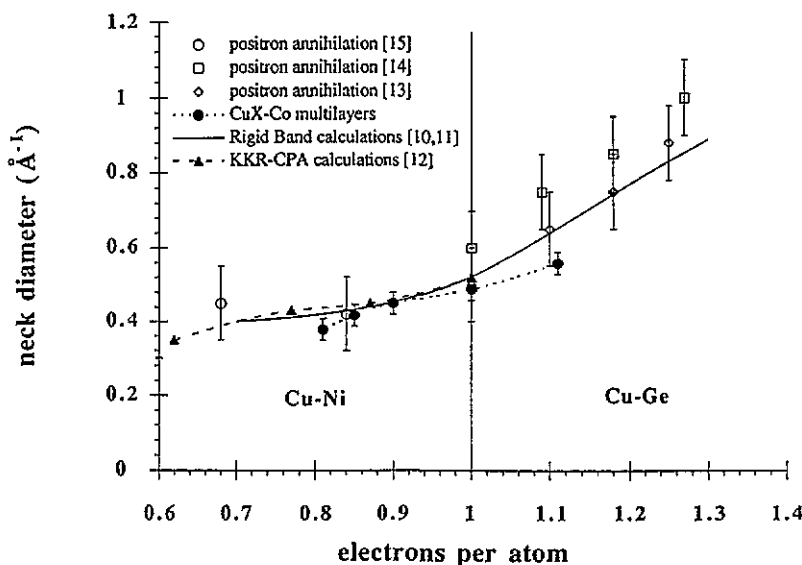

Figure 4. Variations in the neck diameter $q_N = 0.95q_i$ versus the nickel and germanium concentrations which are expressed in electrons per atom. Our data are compared with theoretical predictions and positron annihilation results.

3.2. Antiferromagnetic coupling intensity

The interlayer coupling strength J_i can be deduced from the saturation field H_s of the magnetoresistance from

$$J_i = \tfrac{1}{4} M_s t_M H_s \tag{3}$$

where M_s and t_M are respectively the saturation magnetization and the thickness of the cobalt layers. To a first approximation, at the AF peaks of the coupling, the cosine in equation (1) has its maximum value and then J_i is only a function of $1/t^2$. The values of J_i are reported for all our samples at the first and the second AF peaks in figure 5. One can fit them correctly with a $1/t^2$ law (t is the spacer thickness). This result agrees with the RKKY theory in its pseudo-one-dimensional limit [16]. Concerning the very low values of J_i at the second AF period, we must emphasize the fact that interlayer coupling becomes so weak that hysteresis effects influence it. This is the reason why the latter data are rather noisy.
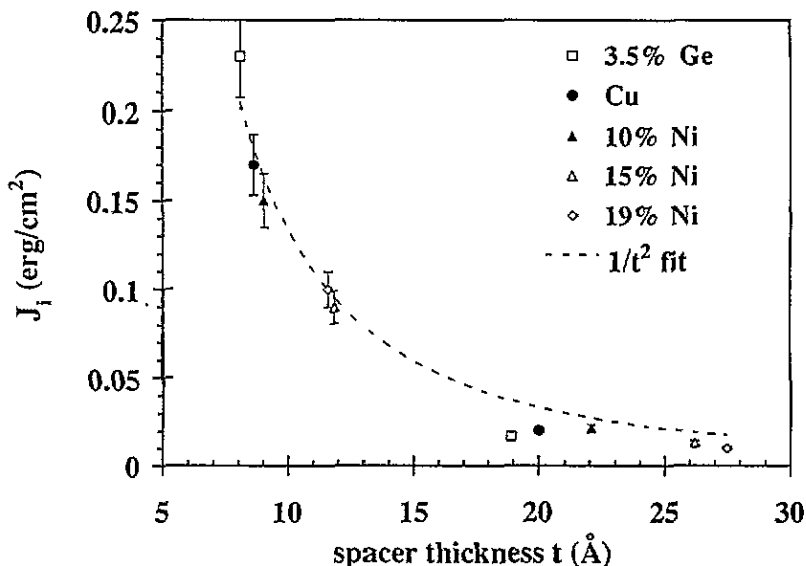

Figure 5. Dependence of the interlayer coupling strength J_i with the spacer layer thickness t at the first and the second AF peaks: ———, $1/t^2$ fit.

From an opposite point of view, if we assume that J_i depends also on the effective mass m^* [4], it should not follow this simple $1/t^2$ rule. In fact, the curvature of the Fermi surface is expected to vary with the amount of impurities. This point has not been clear until now, despite some of the theoretical models developed by, for example, Bruno [17] to describe the mechanism of interlayer coupling better.

3.3. Giant magnetoresistance variations

Disordered dilute alloys have a reduced electronic mean free path λ. Hence, the room-temperature resistivity of the $Cu_{0.81}Ni_{0.19}$ alloy is found to be close to 30 $\mu\Omega$ cm while that of pure copper films does not exceed 10 $\mu\Omega$ cm. This effect is accompanied by a non-metallic behaviour of the temperature dependence of the resistivity for our (Cu–Ni)–Co multilayers with a negative value of $\alpha = (1/R)(dR/dT)$.

This reduction in the mean free path also has repercussions on the GMR, as presented in figure 3; the richer the impurity concentration is in the spacer layer, the faster the GMR decreases from the first to the second AF peak. This dependence of the magnetoresistance
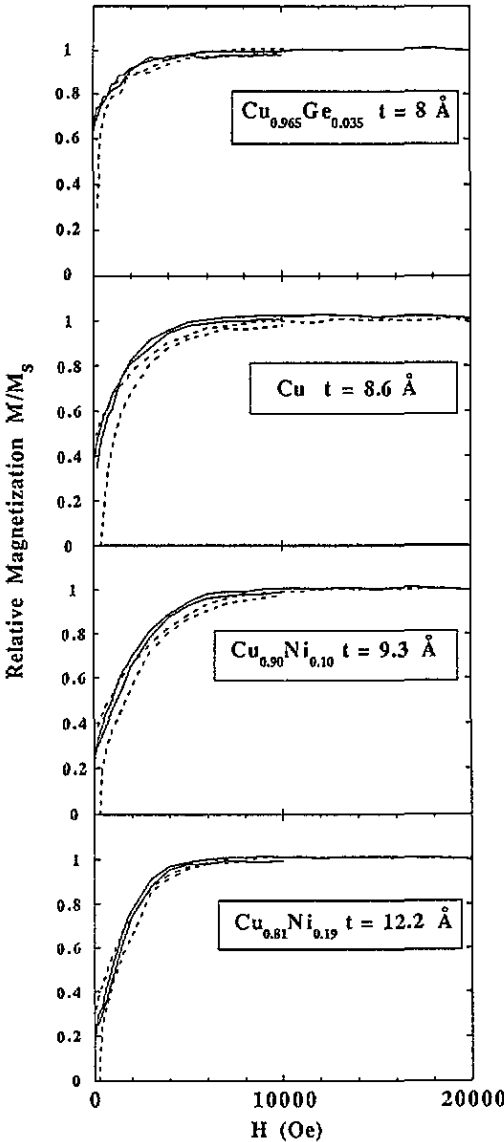
Figure 6. Magnetization curves at the first AF peak of our five alloy multilayers. - - - - $T = 5$ K; —— $T = 300$ K.

allows us to estimate λ, following Bartélémy and Fert [18], if we neglect any conduction through the cobalt layers (if we took it into account, λ would be found to be larger):

$$\Delta R/R = (\Delta R/R_0)\exp(-t/\lambda). \tag{4}$$

This gives reduced values of λ from 17 Å for Cu–Co multilayers down to 7 Å for $Cu_{0.81}Ni_{0.19}$–Co multilayers. Concerning the Cu–Ge-based multilayers, the situation is less clear; the decrease in the GMR at the second peak leads to $\lambda \simeq 15$ Å. In fact, we suppose that the reduction in the mean free path in that case is stronger but masked by incomplete AF coupling at the first GMR peak owing to structural defects of the multilayer. In fact,

the shape of the magnetization curves at the first AF peak for our five alloys confirms this tendency (figure 6); the remanent magnetization increases when the spacer layer thickness is decreased, but the saturation field is not affected too much. This effect, already observed by several workers [3, 9] for low spacer layer thicknesses is now commonly attributed to a loss of the structural integrity of the multilayer stacking and to the presence of local ferromagnetic short circuits, the so-called pinholes [19, 20]. On the contrary, if the coupling had remained perfect at low spacer thicknesses, we would observe a decrease in λ and stronger values of GMR.

If the electronic mean free path was negligible with respect to the periodicity of the multilayers, interlayer coupling which is supported by conduction electrons would be impossible to observe. Fortunately, it is of the same order of magnitude as both of these characteristic lengths, which allows us to obtain our results.

4. Conclusion

We have proved that changes in the electron density of a copper-based spacer layer induce variations in the oscillatory interlayer coupling period for (111)-oriented Co/Cu–Ni (or Ge) multilayers. The correlation of this with the neck diameter of the spacer layer Fermi surface has been clearly established. Such results strongly support the theoretical description of interlayer exchange coupling. Let us emphasize that our results could be obtained because the electronic mean free path in the spacer layer alloy is of the same order of magnitude as the periodicity of the multilayers. This is the main advantage of the use of multilayers for the determination of the neck diameter of resistive alloys. On the contrary, DHVA experiments are unable to give this kind of result because they are more sensitive to impurity scattering. Our data can be compared with positron annihilation results; they are very similar, but with smaller error bars.

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