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Biquadratic magnetic coupling in NiFe/Ag multilayers

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Magnetoresistance, magnetization, and polarized neutron experiments have been performed on $Ni_{81}Fe_{19}$/Ag multilayers. The results give evidence for both biquadratic and bilinear contributions to the total coupling energy. The strong temperature dependence of the biquadratic term leads to a crossover from a canted state at low temperature to an antiferromagnetic one beyond 100 K. We suggest that the biquadratic term could arise from local concentration fluctuations in the chemically disordered NiFe layers.

The study of magnetic coupling in multilayered structures has been a very active field in the last few years. The discovery of antiferromagnetic coupling between magnetic layers and the associated giant magnetoresistance (MR) effects in Fe/Cr multilayers,[1-3] as well as the observation of the oscillatory behavior of the coupling as a function of the thickness of the nonmagnetic layer[4] have resulted in a large amount of both experimental and theoretical work.

More recently[5] it has been reported that, for thicknesses of the spacer layer where the strength of the antiferromagnetic coupling becomes very small, the exchange fluctuations could stabilize another kind of arrangement. In this case the magnetic moments in a given layer tend to lie at 90° to those in the adjacent layer, as expected if a supplementary biquadratic B term is added to the conventional bilinear one in the coupling energy.[5-8] The origin of such a biquadratic term is not yet established, and could be either fundamental (spin-orbit coupling[5]) or purely phenomenological as a result of competing ferromagnetic and antiferromagnetic interactions induced by the presence of steps at the interfaces.

We have recently investigated the magnetic and magnetoresistive properties of $Ni_{81}Fe_{19}$/Ag multilayers prepared by sputtering.[9] Contrary to the case of Ni/Ag multilayers,[10] we observed in the present system nonlinear magnetization curves, with a nonzero magnetic contribution in zero field at low temperature.

In this paper, we show from a detailed study of a sample whose Ag thickness corresponds to the maximum MR ratio that the variations of magnetoresistance, magnetization, and neutron intensities with applied field and temperature completely agree with a model including both bilinear and biquadratic contributions to the total energy. Thanks to the very small anisotropy of $Ni_{81}Fe_{19}$, the values of both coupling constants can be determined separately, even at high temperature where the absolute value of the bilinear term is much larger than that of the biquadratic one. This is a major difference with previous investigations[5-8] where the individual constants could only be evaluated in a much narrower range (i.e., when the equilibrium state in zero field is a canted one). We show that the bilinear coupling constant is essentially independent of temperature. Owing to the strong temperature dependence of the biquadratic term, the equilibrium configuration in zero field changes from a canted state at low temperature to an antiferromagnetic one above 100 K.

The 0.5-μm thick samples were prepared by dc sputtering, by depositing sequentially Ag and $Ni_{81}Fe_{19}$ layers, respectively, 1.08 and 1.22 nm thick, onto glass substrates kept at a temperature of 100 K. The polarized neutron experiments were carried out on 4F1 instrument at Laboratoire Léon Brillouin in Saclay. The magnetization curves were measured using a superconducting quantum interference device (SQUID) magnetometer between 4.2 and 300 K, and the magnetoresistance curves were recorded with a standard four-probe method, the magnetic field being applied perpendicular to the current flow. In all these experiments the magnetic field was applied parallel to the plane of the layers.

Let us consider the magnetic configuration where the spontaneous magnetizations lie in the plane of the layers and, in the absence of any anisotropy, alternately make an angle $+\theta$ and $-\theta$ with the external field. The total energy can be written as

$$E = JM_s^2(1-\cos 2\theta) + BM_s^2(1-\cos 4\theta) - HM_s t\cos\theta \ , \quad (1)$$

where J and B are the bilinear and biquadratic coupling constants, respectively, M_s is the (temperature dependent) spontaneous magnetization in every NiFe layer with thickness t. Such an expression is different from the one usually presented in which the JM_s^2 term is replaced by a unique coefficient A which takes into account both

the magnetization of the layer which polarizes the electron cloud of the nonmagnetic spacer and that of the next magnetic layer to which it is coupled. As is classically written for ferromagnetic or antiferromagnetic substances, relation (1) allows one to separate the thermal dependence of the susceptibility of the coupling electrons from the one of the spontaneous magnetization of the layers.

In zero field, the energy minimum with respect to $\cos\theta$ (B is supposed to be negative) corresponds to $\theta=90°$ when $J/4B>1$ (antiparallel state) and to $\theta=0$ when $J/4B<-1$ (parallel state), whereas a canted state is stabilized for $-1<J/4B<1$, with a crossover similar to that observed in second-order transitions. In the canted state, the equilibrium angle θ_0 is given by $\cos2\theta_0=-J/4B$. For an applied field smaller than the saturation field H_s, the minimization of the energy leads to the following relation:

$$H=(4M_s/t)[(4B-J)\cos\theta-8B\cos^3\theta]\ . \qquad (2)$$

From this relation one can thus extract the field dependence of the net magnetization $M=M_s\cos\theta$ for $H<H_s$ and compare it to the experimentally observed one. Figure 1 shows the magnetization curves recorded at three different temperatures. As we reported previously,[9] the main features of these curves are that the magnetization does not vary linearly with the field below H_s and that there is a ferromagnetic component at zero field for temperatures below 100 K. Such a behavior is somewhat unexpected by reference to our previous studies on Ni/Ag multilayers, where magnetization curves were found in perfect agreement with classical models of antiferromagnetism.[10] Permalloy being even less anisotropic than pure nickel, the observation of a ferromagnetic contribution at zero field cannot be attributed to the coexistence of large ferromagnetic and antiferromagnetic regions, since in this case one would observe a subsequent linear variation of the magnetization with the applied field, contrary to the results presented in Fig. 1. It appears that these results can be well accounted for by considering a supplementary biquadratic term in the total energy, as is clearly demonstrated by the quality of the fits obtained from relation (2) and presented in Fig. 1 as continuous lines.

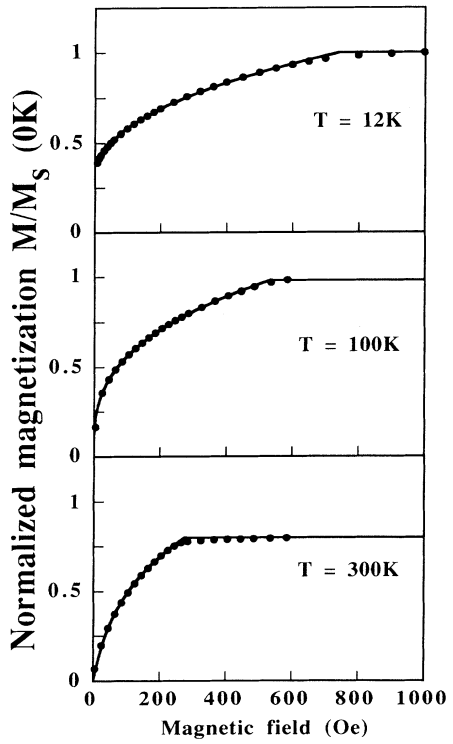

FIG. 1. Magnetization curves for a $NiFe_{1.22}/Ag_{1.08}$ multilayer. Data are normalized to the saturation value at low temperature. The solid lines correspond to a fit to Eq. (2).

Further support to this interpretation is obtained from a study of the field variations of both magnetization and magnetoresistance. Since the MR ratio $\Delta R/R$ is known to scale with the cosine of the angle between magnetizations[10-12] that is with $\cos^2\theta$, one can write, supposing that the initial angle $2\theta_0$ between the magnetizations is different from 180°,

$$\Delta R/R=(R-R_s)/R_s$$
$$=(R_0-R_s)/R_s(1-\cos^2\theta)/(1-\cos^2\theta_0)$$

the constants being determined by the limiting values $R=R_0$ for $\theta=\theta_0$ ($H=0$) and $R=R_s$ for $\theta=0$ ($H=H_s$). Then two cases can be considered. In the first one, we suppose that the sample is inhomogeneous on a macroscopic scale so that the ferromagnetically coupled regions (with magnetization M_0) have a size larger than the electron mean free path, and thus do not participate in the MR effect, whereas the magnetizations in the antiferromagnetically coupled regions are antiparallel in zero field. Thus one has $\cos\theta=(M-M_0)/(M_s-M_0)$ and $\cos\theta_0=0$, leading to

$$\Delta R/R=(R_0-R_s)/R_s[1-(M-M_0)^2/(M_s-M_0)^2]\ . \qquad (3)$$

In the second case, for a homogeneous sample in which the magnetizations are at an angle $2\theta_0$ in zero field, one will have $\cos\theta=M/M_s$ and $\cos\theta_0=M_0/M_s$, leading to

$$\Delta R/R=(R_0-R_s)/R_s(M_s^2-M^2)/(M_s^2-M_0^2)\ . \qquad (4)$$

Both relations are plotted in Fig. 2 for $T=12$ K and

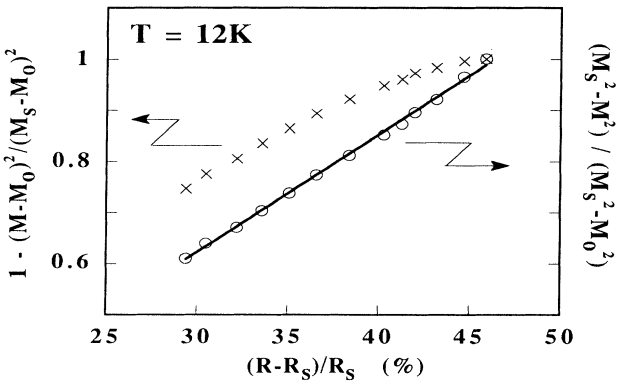

FIG. 2. Variation at 12 K of $1-(M-M_0)^2/(M_s-M_0)^2$ (crosses) and $(M_s^2-M^2)/(M_s^2-M_0^2)$ (circles) with the MR ratio $\Delta R/R$ for $M/M_s<0.7$.

$M/M_s < 0.7$ that is in the region where the influence of M_0 is the most significant. It is evident from this figure that relation (4) is well obeyed, whereas relation (3) is not at all verified. Thus these results clearly show that both the observed nonlinear magnetization curves and the presence of a zero-field contribution are the result of competing ferromagnetic and antiferromagnetic interactions at a microscopic scale.

The neutron scattering technique has been used to check the long-range magnetic order in these multilayers. As was the case for Ni/Ag,[13] a supplementary low angle diffraction peak was observed in zero field at a scattering vector corresponding to twice the chemical periodicity. Let us again consider the ideal case of a homogeneous monodomain system where the magnetizations in successive layers are alternately at $+\theta$ and $-\theta$ from the field direction. The expected polarized neutron intensities defined with respect to the two polarization states ($+$ and $-$) before and after scattering at the sample (I^{++} and I^{--} at a scattering vector $q_1 = 2\pi/\Lambda$, where Λ is the period of the multilayer, and I^{+-} at a scattering vector $q_{1/2} = q_1/2$) are given by

$$I^{++}_{--} = \alpha(\Delta Nb \pm N_m p_m \cos\theta)^2$$

and

$$I^{+-} = \beta(N_m p_m \sin\theta)^2 ,$$

where $\Delta Nb = N_{Ag} b_{Ag} - N_m b_m$ is the nuclear contrast, N_{Ag} and N_m are the silver and nickel-iron atomic densities, b_{Ag} and b_m are the nuclear scattering lengths, p_m is the magnetic scattering amplitude of the NiFe layer and α and β are geometrical coefficients. The intensities are calculated in the kinematic approximation.

The I^{++}, I^{--}, and I^{+-} intensities, normalized at $H=0$, are presented in Fig. 3. Let us first note that the difference $(I^{++} - I^{--})$ is directly related to the value of $\cos\theta$. This confirms that $\cos\theta_0$ is different from zero below 100 K. From the constants J and B extracted from the fit of the magnetization curves, we then calculate the field dependence of the neutron intensities, with p_m as the only adjustable parameter. One sees that a very good agreement with the experimental data is obtained, the fitted p_m values ($p_m = 0.27\mu_{NiFe}$) leading to magnetic moments of 1.01, 1.01, and $0.86\mu_B$ at 12, 100, and 300 K, respectively. These values compare well with both the bulk low-temperature value of $1.04\mu_B$ for $Ni_{81}Fe_{19}$ and the thermal dependence measured from the magnetization curves.

The observation of a magnetic superstructure peak at $q_{1/2}$ implies the existence of an antiferromagnetic component with long-range order. Since the magnetizations in successive layers are not antiparallel, such an order must be stabilized by some kind of next-nearest-neighbor (NNN) ferromagnetic interactions, unless the magnetic coherence is very rapidly lost. Recent nonpolarized neutron experiments on Fe/Ir superlattices seem to support such an interpretation.[14] In the case of NiFe/Ag multilayers, we observed an influence of the magnetic field on

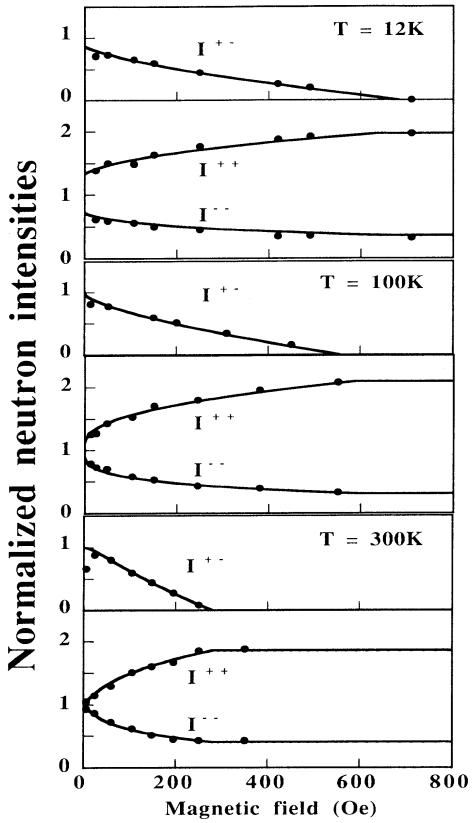

FIG. 3. Polarized neutron intensities I^{++}, I^{--} and I^{+-} as a function of applied field for the same sample as in Fig. 1. The solid lines are fits obtained with p_m as the only adjustable parameter.

the width of the magnetic diffraction peak (i.e., on the magnetic coherence length) for temperatures smaller than 100 K that is in the canted state. This could indicate that different kinds of magnetic structures can be stabilized in such systems, depending on the sign and magnitude of the NNN interactions.[5]

Figure 4 gives the temperature dependence of the coupling constants J and $4B$ extracted from the fit of the magnetization curves. As was already apparent from Figs. 1 and 3, the ratio $J/4B$ progressively increases with

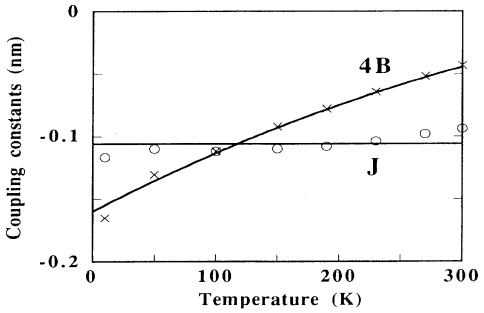

FIG. 4. Temperature dependence of the coupling constants J and $4B$. The constant B is fitted to $B = B_0(1 - T/T_0)^2$ with $T_0 = 630$ K.

temperature from about 0.7 at 10 K to 2.2 at room temperature. In other words, the equilibrium angle between magnetizations in zero field is about 135° at 10 K and reaches 180° beyond 100 K.

Concerning the coupling constants J and B, it appears that their respective temperature dependences are very different. First the J constant is found almost temperature independent. We also observed the same behavior in our previous investigations of Ni/Ag multilayers,[10] provided the total energy is written according to Eq. (1). On the contrary, the absolute value of the B constant decreases very rapidly with temperature, and can be well fitted to $B=B_0(1-T/T_0)^2$, with $T_0=630$ K. Such a behavior has been evidenced in Fe/Al/Fe trilayers.[7] Converting to energy units, JM_s^2 varies from -6.0×10^{-6} J/m^2 at 10 K to -3.8×10^{-6} J/m^2 at 300 K, whereas BM_s^2 goes from -2.2×10^{-6} to -0.4×10^{-6} J/m^2.

The occurrence of a biquadratic contribution is generally attributed to the presence of interface steps, leading to a competition between ferromagnetic and antiferromagnetic interactions.[5,15] Although this possibility cannot be eliminated in the present NiFe/Ag system, we point out that alloying effects could be responsible for such a magnetic behavior. Indeed it has been observed recently[16] that, in NiFeCo/Cu multilayers, the position of the first MR peak was dependent on the composition of the magnetic alloy layer, going from a Cu thickness of 0.9 nm for $Ni_{40}Fe_{60}$ to 1.11 nm for pure nickel. We also observed the same trend in Ag/NiFe multilayers. Since the position of the MR peak is related to the critical thickness for antiferromagnetic coupling, one can imagine that, as long as the magnetic alloy is not chemically ordered, different local atomic concentrations in the magnetic layers on both sides of the (ideally flat) nonmagnetic one will lead to different values of the coupling (in both sign and magnitude), and thus to competing interactions at a microscopic scale.

In conclusion, we have presented in this paper an example of a multilayered system with both bilinear and biquadratic contributions to the coupling energy. The validity of the model is confirmed by the variation of the MR ratio and magnetization with the field at low temperature. Very good fits of both magnetization and polarized neutron results are obtained on the whole field and temperature ranges. They allow the determination of the coupling constants even in the case where the equilibrium configuration in zero field is an antiferromagnetic one. The bilinear coupling constant is found practically independent of temperature, in contrast to the strong temperature variation of the biquadratic one.

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