Biquadratic coupling in sputtered Fe/Cr/Fe still in need of a new mechanism

S. M. Rezende,[a] C. Chesman, M. A. Lucena, M. C. de Moura, A. Azevedo, and F. M. de Aguiar
Departamento de Física, Universidade Federal de Pernambuco, Recife PE 50670-901, Brazil

S. S. P. Parkin
IBM Almaden Research Center, San Jose, California 95120-6099

The bilinear (J_1) and biquadratic (J_2) exchange coupling constants were measured in sputtered trilayers of (100) Fe(40 Å)/Cr(s)/Fe(40 Å) for several Cr spacer layer thicknesses in the range $s=8$–35 Å and as a function of temperature T, using magneto-optical Kerr effect magnetometry, Brillouin light scattering, and ferromagnetic resonance. In the samples in the range $s=8$–13 Å, corresponding to the first antiferromagnetic peak of J_1, J_2 follows J_1 with a room temperature ratio $J_2/J_1 \cong 0.1$, while in the range 25–35 Å, corresponding to the second antiferromagnetic peak, J_2 also follows J_1 but with a much larger ratio $J_2/J_1 \cong 1$. This result, as well as the temperature dependence of J_2 in all samples but the one with $s=15$ Å, cannot be explained by any of the intrinsic or extrinsic mechanisms that have been proposed for the origin of the biquadratic exchange coupling in Fe/Cr/Fe. © *1999 American Institute of Physics.* [S0021-8979(99)68008-2]

INTRODUCTION

Magnetic multilayers, consisting of stacks of ferromagnetic layers separated by nonmagnetic metallic layers, have attracted considerable attention due to their unique physical properties and potential for technological applications. Many multilayer systems exhibit a coupling between the magnetic layers mediated by the nonmagnetic spacers, which oscillates periodically between ferromagnetic (FM) and antiferromagnetic (AFM) as the spacer-layer thickness varies in the range of 5–50 Å.[1–5] Due to its central role in the properties of magnetic multilayers, the coupling between the magnetic layers through the nonmagnetic metallic spacer has been the subject of extensive investigations for nearly ten years. Experimentally this coupling is more conveniently studied in a trilayer structure, formed by two magnetic thin films separated by a nonmagnetic layer, Fe/Cr/Fe being the most studied system.[4–12]

The coupling between the magnetic layers is usually dominated by a mechanism which can be modeled by an interaction energy of the form $-J_1\mathbf{m}_1\cdot\mathbf{m}_2$, where $\mathbf{m}_1$ and $\mathbf{m}_2$ are the unit magnetizations of the two magnetic layers and J_1 is the bilinear exchange coupling constant. The origin of this coupling lies in the interaction between the s electrons in the metallic spacer and the d electrons in the magnetic layers,[13–15] and is currently well understood.[16] However, more recently it was observed[5,6,17] that under certain conditions the magnetic moments of the two layers tend to align at 90° with respect to each other. This alignment may be accounted for through an interaction energy described by a phenomenological biquadratic exchange coupling (BEC) $-J_2(\mathbf{m}_1\cdot\mathbf{m}_2)^2$, where J_2 is the biquadratic coupling constant. Over the last few years several mechanisms have been proposed for the origin of the biquadratic coupling,[18–21] some attributed to intrinsic properties of the spacer layer and others to extrinsic factors, such as the presence of impurities or interface roughness. Intrinsic mechanisms seem not to account for the observed coupling strengths and signs, nor for its temperature dependence.[18] On the other hand, if the mechanisms based on extrinsic factors prevails in the BEC, one expects the value of the coupling constant J_2 to be quite sensitive to details of the sample preparation conditions. Indeed, there is a considerable spread in the values of J_2 measured by different groups for nominally the same system, indicating the dominance of extrinsic mechanisms. However, there are conflicting reports on the temperature and spacer layer thickness dependence of J_2 and only in very few cases there seems to be a reasonable connection between data and some specific extrinsic mechanism.[22] The fact is that the whole question of the origin of the BEC is still quite controversial[23] and deserves further investigation. In this article we present new data on the temperature dependence of J_1 and J_2 in the prototype system (100) Fe/Cr/Fe for varying Cr spacer layer thickness and show that in only one sample the exchange fluctuation mechanism accounts for the experimental data.

EXPERIMENTAL RESULTS

The samples investigated are single-crystal trilayer structures of (100) Fe(40 Å)/Cr(s)/Fe(40 Å) grown by magnetron sputter deposition, as described in Ref. 25 on MgO (100) substrates. All samples have the same Fe layer thickness, $d=40$ Å, and a thin Cr cap layer. Initial characterization of the coupling was made by magneto-optical Kerr effect (MOKE) with a Cr wedged sample, with $0<s<70$ Å. Then a series of samples was prepared with uniform Cr thickness varying from 5 to 35 Å, a range that corresponds to the first two antiferromagnetic peaks.

[a]Electronic mail: smr@df.ufpe.br

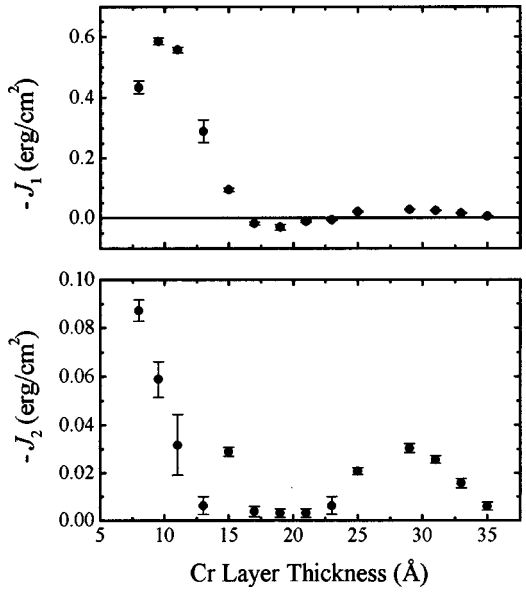

FIG. 1. Room-temperature exchange coupling constants measured in sputtered (100) Fe(40 Å)/Cr(s)/Fe(40 Å) by MOKE, BLS, and FMR.

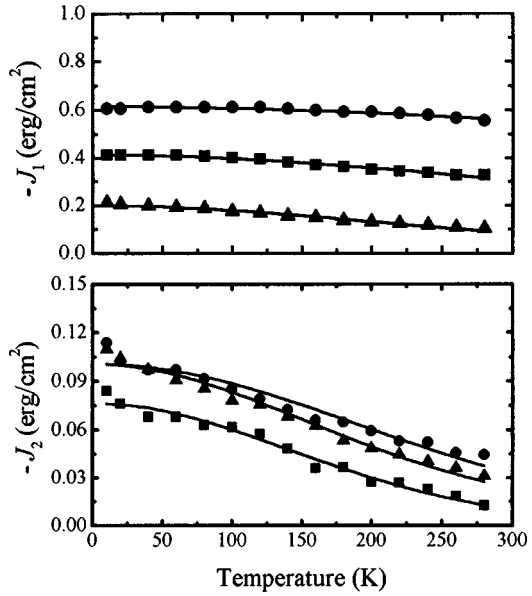

FIG. 2. Temperature dependence of the exchange coupling constants in Fe(40 Å)/Cr(s)/Fe(40 Å). The symbols represent the data for the samples with $s=11$ (circles), 13 (squares), and 15 Å (triangles) and the solid lines are fits with theoretical predictions.

In order to obtain reliable values for J_1 and J_2, we have used three independent techniques, namely, MOKE magnetometry, Brillouin light scattering (BLS), and ferromagnetic resonance (FMR). The data were fitted with a phenomenological energy model including bilinear and biquadratic exchange couplings, as well as surface and crystalline cubic anisotropy contributions. Details of the measuring techniques and the procedures used to extract the values of J_1 and J_2 are presented elsewhere.[24,25]

Figure 1 shows the room-temperature values for J_1 and J_2measured in 14 samples with varying Cr spacer thickness. The vertical bars represent the uncertainties due to the estimated errors in each fitting plus the spread in the values obtained with the various techniques. Two AF peaks alternating with one FM peak are observed in J_1 in the thickness range 5 Å$<s<$35 Å, a well known result which has been obtained by many authors. The maximum (absolute) value of J_1 in the first AF peak is 0.59 erg/cm^2, for $s=9.5$ Å, a value somewhat smaller than $J_1 \cong 1$ erg/cm^2 reported for some molecular beam epitaxy (MBE) grown samples,[3,4,9] but similar to those reported for other MBE[7] and sputtered[11] (100) Fe/Cr/Fe trilayers.

The result for J_2 is not so well known. In fact, to our knowledge, this is the first measurement of J_2 vs spacer-layer thickness in the second AF peak. The data show that J_2 is negative in the whole range, and that its ratio to J_1 varies considerably with s. In most of the first AF peak, J_2 follows a dependence with s similar to that of J_1, with $J_2/J_1 \cong 0.1$. However, near the crossing from AF to FM ($s=15$ Å), this ratio increases to $J_2/J_1 \cong 0.3$, which is similar to that measured in a structure Fe(28 Å)/Cr(15.8 Å) grown by MBE on a (100) Fe whisker.[8] Throughout the Cr thickness range $s=16$–24 Å, corresponding to the second FM peak, the ratio $-J_2/J_1$ remains in the range of 0.2–0.3. Surprisingly, in the second AF peak the ratio increases to $J_2/J_1 \cong 1$, so that the antiferromagnetic phase ceases to exist.[24,25]

Figure 2 shows the temperature dependence of J_1 and J_2 measured in three samples with Cr layer thickness $s=11$, 13, and 15 Å. Qualitatively the data are similar to results previously obtained in several systems, both exchange constants decrease with increasing temperature. However, a detailed analysis of the temperature dependence contains important clues on the mechanisms responsible for the coupling between the magnetic layers.

DISCUSSION

In order to discuss the origin of the biquadratic coupling in our Fe/Cr/Fe samples, we start looking at the behavior of the bilinear coupling J_1. There is general agreement today that the bilinear coupling originates in the interaction between the s electrons in the Cr layer and the d electrons in the Fe layers, the so-called intrinsic mechanism. Calculations taking into account the full electronic structure of the metals show[26,27] that for perfectly sharp interfaces the behavior of J_1 with the spacer layer thickness is entirely dominated by short period oscillations with amplitude decaying with increasing thickness. The maximum negative value of $J_1$is approximately 7 erg/cm^2, which is an order of magnitude larger than the measured values. This discrepancy is accounted for by the existence of roughness, interdiffusion, vacancies, and steps in the real sample, which smooth out the short period oscillations and drastically reduce the peak value.[27] While comparison between theory and experimental data for the strength of the coupling is not satisfactory, the same is not true for the temperature dependence of J_1. Consider the theoretical prediction for the intrinsic mechanism[21] $J_1(T) = J_1(0) f_1(T)$, where $f_1(T) = (T/T_0)/\sinh(T/T_0)$.

The solid lines in Fig. 2(a) represents the fits of this function to the experimental data, obtained with $T_0=390$, 214, and 122 K for the samples with Cr layer thickness

$s=11$, 13, and 15 Å, respectively. Note that T_0 decreases with increasing s, and although it does not follow the $1/s$ law of the simple theory, the good fits indicate that the intrinsic mechanism accounts for the origin of the bilinear exchange coupling.

Regarding the origin of the biquadratic coupling J_2, we first note that it cannot be attributed to intrinsic mechanisms for two reasons: the predicted oscillation period for J_2 is smaller than for J_1, whereas the data of Fig. 1 shows J_2 following J_1; theory[21] predicts a rapid decay of J_2 with increasing s, which is certainly not the case of the data. For the samples under investigation here, among the various extrinsic sources proposed for J_2, the most plausible one is the Slonczewski's exchange fluctuation mechanism[18] caused by interface roughness. According to the model, J_2 arises from the combined effect of the rapid oscillation in the intrinsic J_1 and the variation in spacer layer thickness in the form of terraces. If J_1 varies in steps of $\pm 2\Delta J_1$, the first order contribution of this mechanism to the BEC is[18]

$$J_2 = -\frac{4(\Delta J_1)^2 L}{\pi^3 A}\coth\left(\frac{\pi d}{L}\right), \tag{1}$$

where A is the exchange stiffness constant of the Fe layer, L is the terrace width, and d is the Fe layer thickness. Equation (1) predicts that J_2 is always negative, favoring the 90° alignment, as observed in the experiments, and that its strength varies with the square of ΔJ_1. Considering that ΔJ_1 is a step change in the bilinear coupling arising from the short period oscillation, and that the measured coupling represents an average of J_1, Eq. (1) predicts for J_2 a temperature dependence following $J_1^2(T)/A(T)$. In order to verify this prediction, it is necessary to take into account the temperature variation of the exchange stiffness.[23] Thus we have determined $A(T)$ by measuring the volume mode frequencies in a 250 Å thick single film of (100) Fe/MgO as a function of temperature using BLS.

We now argue that the present model of the exchange fluctuation mechanism cannot by itself explain the measured BEC in the whole range of Cr spacer layer thickness. The first argument is that Eq. (1) predicts that the amplitude of J_2 decays with increasing s following J_1^2. This is in complete disagreement with the data, which show that while the peak amplitude of J_1 does decrease with increasing s, the ratio J_2/J_1 is $\cong 0.1$ in the first AF peak and $\cong 1$ in the second AF peak. The second argument is based on the temperature dependence of the coupling constants. The solid lines in Fig. 2(b) are the fits of the $J_2(T)$ data with $[f_1(T)]^b$, obtained with the values $b=11.8$, 6.6, and 1.7 for the samples with Cr layer thickness $s=11$, 13, and 15 Å, respectively. This shows that only for the sample with $s=15$ Å the temperature variation of J_2 is close to the $J_1^2(T)$ dependence predicted by the exchange fluctuation mechanism. In order to verify if this dependence really applies to the $s=15$ Å sample, one needs to take into account the temperature variation of the exchange stiffness. So we fitted the BLS data with $A(T) \propto [f_1(T)]^a$,, with $T_0=122$ K appropriate for this sample. This yields an exponent $a=0.25\pm 0.10$, implying that the temperature dependence of J_2 for the $s=15$ Å sample is consistent with the prediction of the exchange fluctuation mechanism. However none of the proposed mechanisms for the BEC[12,18] can account quantitatively for the data in the other Fe/Cr/Fe samples. Therefore, the present results add evidence to previous[12,23] conclusions that further theoretical and experimental work is necessary to fully explain the biquadratic exchange coupling in magnetic multilayers.

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