Analysis of bilinear and biquadratic exchange coupling in Fe/Ag/Fe(001) trilayers

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Ferromagnetic resonance (FMR) and surface magneto-optical Kerr effect (SMOKE) studies of the exchange coupling in Fe/Ag/Fe(001) structures are presented. The interfaces in these structures can be improved significantly by growing the first Fe(001) layer at a raised substrate temperature. The exchange couplings in Fe/Ag/Fe trilayers were studied as a function of the interlayer thickness and temperature. The improved interfaces in the Fe/Ag/Fe system revealed new features in the exchange coupling which were absent in samples grown entirely at room temperature. Quantitative data from the FMR and SMOKE measurements are compared. The magnetization loops for Fe/Ag/Fe trilayers can be explained well only by including the simultaneous presence of bilinear and biquadratic exchange coupling. It is shown that the exchange coupling through Ag(001) exhibits long wavelength oscillations.

The aim of our studies is to investigate the influence of interface roughness on the exchange coupling between two ferromagnetic layers coupled through a nonferromagnetic interlayer. The role of interface roughness has been recently treated by Slonczewski.[1] Slonczewski showed that when the exchange interaction changes rapidly with the number of monolayers, and when the interface consists of randomly distributed atomic terraces, then an additional term $-J_1 \sin^2 \Theta$ arises in the effective exchange interaction energy, E, which couples two ferromagnetic layers through a nonmagnetic interlayer

$$E = -J_0 \cos \Theta - J_1 \sin^2 \Theta, \tag{1}$$

where Θ is the angle between the magnetic moments. J_1, known as the biquadratic exchange energy, is always positive.

We have recently reported studies of the exchange coupling in Fe(001)/Cu, Pd, Ag, Au(001)/Fe(001) structures[2] grown at room temperature (RT). The interface at the Fe(001) layers in those structures possessed a roughness which is limited to at most two atomic layers. The growth process of Fe layers on Ag(001) proceeds in a quasi-layer-by-layer mode with nucleation of new atomic clusters 1-monolayer (ML) thick on top of the unfinished layer. Those clusters maintain approximately an equidistant separation of the order of 60 Å which was determined from the splitting of reflection high energy electron diffraction (RHEED) streaks.[3] On the other hand the top surface of nonmagnetic interlayers in Fe/Ag, Cu, Pd/Fe trilayers do not maintain the same roughness as the first Fe layer. The absence of RHEED streak splitting (after only one additional atomic layer is deposited on the top of the Fe(001) layer) and the sharpness of the RHEED patterns indicate that the average atomic terrace width is significantly larger than that obtained for Fe grown on Ag(001) at RT. The intention of this paper is to present the results of our studies of the exchange coupling in structures in which the first Fe layer possessed an interface characterized by large atomic terraces comparable to that of the Ag(001) interlayers.

Our studies of the exchange coupling were carried out using ferromagnetic resonance (FMR) and surface magneto-optical Kerr effect (SMOKE) techniques. The merits of both techniques and the interpretation of the measured data have been presented in our recent paper.[4] The measured magnetization loops did not show a visible hysteretic behavior and therefore the analyses were carried out by using the minimum energy path approach. The trilayers were prepared in a PHI-400 molecular beam epitaxy machine equipped for RHEED and Auger electron spectroscopy. All the structures studied were grown on well-prepared Ag(001) single crystal substrates.[5]

In order to increase the smoothness of the first Fe layer on Ag(001) we divided the Fe growth process into two steps.[6] The first 5–6 ML of Fe were grown at RT to protect the Ag/Fe interface from atomic intermixing. Then the substrate temperature was raised to ~150 °C and an additional 3–4 ML of Fe were deposited. The RHEED streak splitting disappeared after the deposition of the equivalent of an additional ~1 ML of Fe and the amplitude of the RHEED intensity oscillations increased substantially, ~two-to threefold, compared with those observed during the growth at RT. For the Fe(001) layers prepared in the two-stage process the corresponding RHEED patterns were sharp and comparable with the patterns observed using bare Ag(001) substrates. This indicated that the average terrace width was of the order of 400 Å. The Ag interlayer and the second Fe film, usually 16 ML thick, were deposited at RT. All trilayers were protected by 20 ML of epitaxial Au before being exposed to ambient conditions for the magnetic measurements.

The FMR technique determines the exchange coupling between the two magnetic layers for a configuration in which the magnetizations in the two layers are nearly parallel; it therefore provides a measure of $J^{AB} = (J_0 - 2J_1)$, see Eq. (1). It is useful to define the parameter J^{AB} as the total exchange coupling parameter.

In Fe/Ag/Fe trilayers with the first Fe layer grown at elevated temperatures, the thickness dependence of the exchange coupling is different from that observed in samples which were entirely prepared at RT, see Fig. 1. In Fe/

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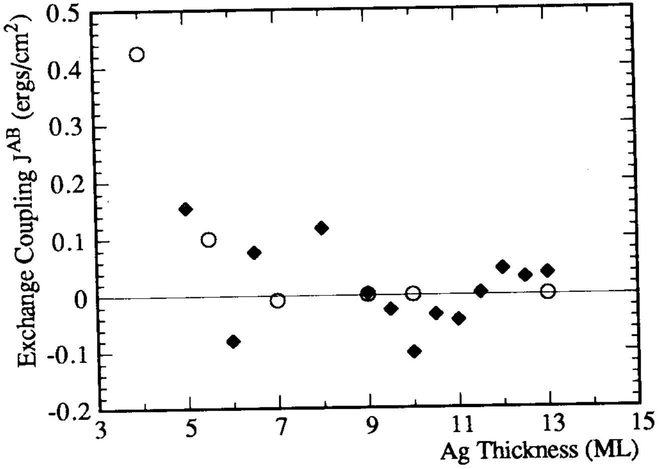

FIG. 1. The thickness dependence of the total exchange coupling parameter $J^{AB}=J_0-2J_1$ observed for Fe/Ag/Fe trilayers measured at 295 K. Closed symbols correspond to trilayers with the first Fe layer grown at an elevated temperature; open symbols correspond to samples grown entirely at RT.

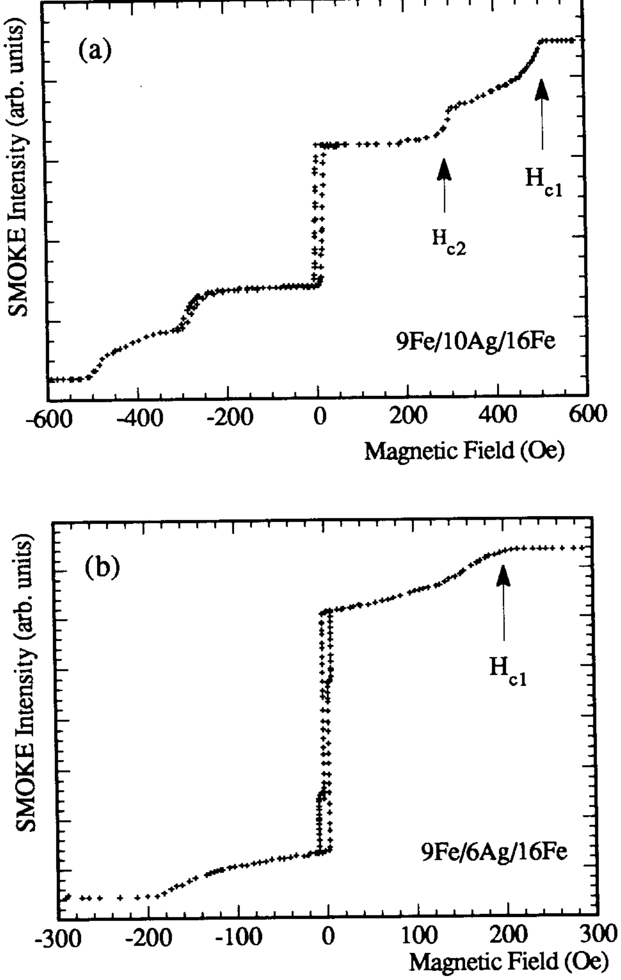

FIG. 3. Hysteresis loops measured using the SMOKE technique. The applied field lies along an easy magnetic axis {100}. (a) Sample 9Fe/10Ag/16Fe, (b) sample 9Fe/6Ag/16Fe.

Ag/Fe trilayers entirely grown at RT the ferromagnetic exchange coupling decreased rapidly with increasing interlayer thickness. It reached zero at ~7 ML and the two iron films remained decoupled for thicker interlayers.[2] Similar results were observed by Fuss *et al.*[7] On the contrary, the thickness dependence of the exchange coupling in structures grown at elevated temperatures clearly exhibited oscillatory behavior. Maxima of the antiferromagnetic coupling occurred at 6- and 10-ML-thick Ag interlayers, see Figs. 1 and 2. Note that the values of the exchange coupling oscillate around zero. This behavior is different from that of Fe/Cu/Fe trilayers with the first Fe layer grown at elevated temperatures[6] for which the measured exchange coupling, although oscillatory, remains antiferromagnetic for thicknesses greater than 8 ML.

The exchange coupling in Fe/Ag/Fe trilayers entirely prepared at RT is temperature independent.[2] Figure 3 shows that Fe/Ag/Fe trilayers with the first Fe layer grown at elevated temperatures clearly exhibit a temperature dependence.

For Fe/Ag/Fe trilayers in which the FMR measurements exhibited antiferromagnetic coupling, two types of behavior were observed in the SMOKE experiments for the applied field directed along one of the in-plane, fourfold easy {100} axes, see Figs. 3(a) and 3(b). One of them is shown in Fig. 3(a) where two critical fields, H_{c1} and H_{c2}, are clearly visible. H_{c1} corresponds to the field at which the magnetic moments of the individual Fe layers start to deviate from the direction of the dc external field. H_{c2} corresponds to the onset of an antiferromagnetic configuration in which the magnetization of the thicker Fe film lies along the field direction, and the magnetization in the thinner Fe layer approaches the direction opposite the applied field. Figure 3(b) shows an example where H_{c2} is absent altogether. The total magnetic moment in zero applied magnetic field in this case corresponds to a configuration in which the magnetic moments in the two Fe layers are nearly oriented along mutually perpendicular easy axes ({100}) with the thicker Fe layer oriented along the direction of the applied dc magnetic field. It was necessary to

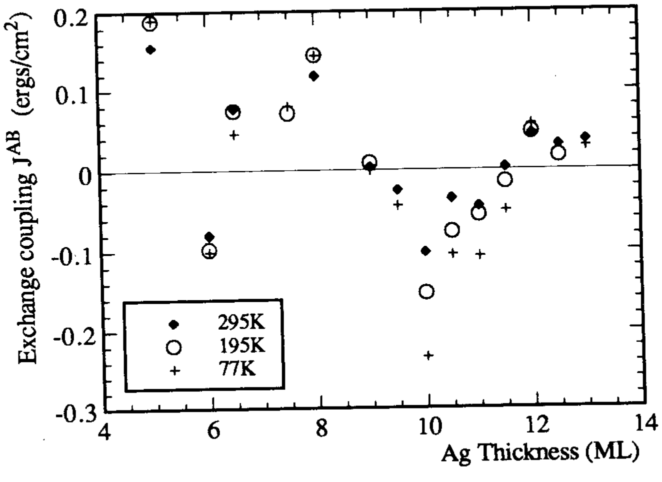

FIG. 2. The thickness dependence of the total exchange coupling parameter $J^{AB}=J_0-2J_1$ measured at 295, 195, and 77 K for Fe/Ag/Fe trilayers with the first Fe layer grown at an elevated temperature.

TABLE I. The results of the exchange coupling as a function of Ag interlayer thickness deduced from SMOKE and FMR measurements at 295 K for antiferromagnetically coupled trilayers. All results are listed in units of ergs/cm^2. J_0 and J_1 denote the bilinear and biquadratic exchange coupling parameters, see Eq. (1), deduced using the path of minimum energy and using the following parameters: $4\pi M_{\rm eff}=6.1$ kOe, $2K_1/M_s=0.32$ kOe for the 9 ML Fe layer; $4\pi M_{\rm eff}=15.5$ kOe, $2K_1/M_s=0.47$ kOe for the 16.6 ML Fe layer. Bilinear minimum denotes $J^{\rm AB}=J_0-2J_1$, calculated from the SMOKE data. FMR results denotes value of the total exchange coupling determined from FMR measurements.

Sample	J_0	J_1	Bilinear minimum	FMR results
9Fe/6Ag/16Fe	0.053	0.070	−0.087	−0.080
9Fe/9.5Ag/16Fe	0.015	0.021	−0.027	−0.025
9Fe/10Ag/16Fe	−0.057	0.032	−0.12	−0.102
9Fe/10.5Ag/16Fe	−0.008	0.026	−0.06	−0.035

use a biquadratic term for the exchange coupling [Eq. (1)] in our calculations in order to reproduce the observed field dependence of the magnetization loops measured by means of SMOKE. The results of our analyses, which yield values of J_0 and J_1, are summarized in Table I.

The presence of a biquadratic exchange coupling in our measurements is most likely caused by Slonczewski's mechanism.[1] The Slonczewski model predicts that in the limit of zero terrace size the contribution to the biquadratic exchange coupling disappears. Therefore trilayers exhibiting a fast spatial variation acquire properties corresponding to the average torque, which in our case translates into the average bilinear exchange coupling. For structures grown entirely at RT the exchange coupling was equal to zero for samples with Ag interlayer thicknesses greater than 7 ML. This surprising result indicates that the bilinear exchange coupling can be effectively removed by averaging over short terraces ($\sim$5–6 nm, at the first Fe/Ag interface) which are faced by large atomic terraces of the second Ag/Fe interface. This suggests that the exchange coupling in Fe/Ag/Fe(001) samples with perfect interfaces oscillates rapidly and very symmetrically around zero. By growing at elevated temperatures we increased the average terrace size at the first Fe/Ag interface and recovered the oscillatory behavior of the exchange coupling. It is interesting to note that one can reproduce the value of the exchange coupling much better among samples which were grown entirely at RT compared to the Fe/Ag/Fe samples in which the first Fe layer was grown at an elevated temperature. In samples with large and comparable atomic terraces (at the Fe/Ag and Ag/Fe interfaces) one can expect a contribution to the exchange coupling from the short wavelength oscillatory component [$\lambda=2.4$ ML in Ag(001)]. The local interlayer thickness is given by the distance between atomic terraces facing each other. The contribution of the short wavelength oscillations depends on the degree to which atomic terraces are cross correleted to each other. A completely statistical distribution removes entirely the contribution of the short wavelength oscillations. For samples with large and comparable terraces one can expect some degree of correlation between terraces and therefore some contribution of the short wavelength oscillation to the exchange coupling. Small changes in the growth conditions, and/or our ability to reproduce a given thickness within $\sim$0.2 ML, most likely resulted in small displacements between atomic terraces, and consequently leads to a different value of the measured exchange coupling. For example, in two samples each having a 6 ML Ag interlayer the total exchange coupling at 295 K was found to be −0.080 and −0.035 ergs/cm^2, respectively; the error in the determination of the exchange coupling using FMR was estimated to be ± 0.006 ergs/cm^2. Note that the strength of the exchange coupling in Fe/Ag/Fe(001) films is very similar to that in Fe/Cu/Fe(001)[6,8] and Fe whisker/Cr,Cu/Fe(001)[9] samples; therefore the behavior of the exchange coupling through fcc Ag(001) does not deviate from the general behavior which has been observed in other nonmagnetic interlayers.

The thickness dependence of the exchange coupling shows that the spacing corresponding to ferromagnetic coupling and to antiferromagnetic coupling is approximately 2.5–3 ML; therefore the corresponding period of oscillations would be 5–6 ML. This value compares well with the calculated long wavelength period for fcc Ag(001), $\lambda=5.6$ ML. The oscillatory exchange coupling which we observe is in agreement with the sign of the coupling obtained in recent SEMPA studies by the NIST group[10] of wedged samples (Fe whisker/Ag/Fe). In a recent paper Ortega and Himpsel[11] reported observations of the quantum-well states at the Fermi level in Ag grown on bulk Fe(100) using inverse photoemission. Near E_F the observed quantum-well states (having *s-p* character) were spin polarized and exhibited oscillatory behavior with a periodicity of 5 ML. These states may mediate the coupling between two Fe layers separated by an Ag spacer.

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[1] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[2] Z. Celinski and B. Heinrich, J. Magn. Magn. Mater. **99**, L25 (1991).

[3] B. Heinrich, K. B. Urquhart, J. R. Dutcher, J. F. Cochran, A. S. Arrott, D. A. Steigerwald, and W. F. Egelhoff, Jr., J. Appl. Phys. **63**, 3863, (1988).

[4] B. Heinrich, J. Kirschner, M. Kowalewski, J. F. Cochran, Z. Celinski, and A. S. Arrott, Phys. Rev. B **44**, 9348 (1991)

[5] B. Heinrich, S. T. Purcell, J. R. Dutcher, J. F. Cochran, and A. S. Arrott, Phys. Rev. B **38**, 12879 (1988)

[6] B. Heinrich, Z. Celinski, J. F. Cochran, A. S. Arrott, K. Myrtle, and S. T. Purcell, Phys. Rev. B **47**, 5077 (1993).

[7] A. Fuss, S. Demokritov, P. Grünberg, and W. Zinn, J. Magn. Magn. Mater. **103**, L221, (1992).

[8] S. T. Purcell, W. Folkerts, M. T. Johnson, N. W. E. McGee, K. Jager, J. aan de Stegge, W. B. Zeper, W. Hoving, and P. Grünberg, Phys. Rev. Lett. **67**, 903 (1991); M. T. Johnson, S. T. Purcell, N. W. E. McGee, R. Coehoorn, J. aan de Stegge, and W. Hoving, *ibid.* **68**, 2688, (1992).

[9] J. Unguris, and R. J. Cellota, D. T. Pierce, presentation at NATO Advanced Research Workshop on Magnetism and Structure in Systems of Reduced Dimension, Cargese, June 1992 (unpublished).

[10] J. E. Ortega and F. J. Himpsel, Phys. Rev. Lett. **69**, 844, (1992)