REPRINTED FROM:

journal of magnetism and magnetic materials

Journal of Magnetism and Magnetic Materials 127 (1993) 205–213
North-Holland

Oscillatory magnetic coupling in Fe/Ag/Fe(100) sandwich structures

J. Unguris, R.J. Celotta and D.T. Pierce
National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

Received 23 November 1992; in revised form 12 January 1993

NORTH-HOLLAND

Journal of Magnetism and Magnetic Materials 127 (1993) 205–213
North-Holland

journal of magnetism and magnetic materials

Oscillatory magnetic coupling in Fe/Ag/Fe(100) sandwich structures

J. Unguris, R.J. Celotta and D.T. Pierce

National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

Received 23 November 1992; in revised form 12 January 1993

The magnetic coupling in Fe/Ag/Fe sandwich structures has been studied using scanning electron microscopy with polarization analysis (SEMPA). when atomically well-ordered Ag spacer layers are grown epitaxially on an Fe whisker substrate, the coupling between the Fe substrate and the top Fe film is found to oscillate between ferromagnetic and antiferromagnetic as a periodic function of the Ag thickness. The magnetic coupling is composed of two oscillatory components with periods of 2.37 ± 0.07 and 5.73 ± 0.05 Ag layers. The oscillations persist for Ag spacer films that are up to at least 50 layers thick. These periods are consistent with theoretical models that predict that the oscillatory coupling length scale is determined by spanning vectors of the spacer material's Fermi surface. Biquadratic coupling was also observed in these structures. The relative strength of biquadratic to bilinear coupling was found to depend on the thickness of the top Fe film.

1. Introduction

The exchange coupling between magnetic thin films separated by certain nonmagnetic spacer layers is found to oscillate between ferromagnetic (FM) and antiferromagnetic (AFM) as a function of the spacer thickness. Understanding the origin of this oscillatory magnetic coupling is both of intrinsic interest and has important implications for sensor technology, but understanding these magnetic properties requires films that are structurally well characterized. The atomic-scale quality of the film growth critically influences such coupling properties as periodicity, strength, and direction. While nearly all exchange coupled polycrystalline films seem to show about the same oscillation period, about 1 nm [1], atomically well-ordered, single-crystal epitaxial films exhibit more complex behaviors. In addition to the long-period oscillations, short-period oscillations with a period of a few atomic layers have also been observed for Fe films separated by Cr [2–4], Mn [5], Au [6], Mo [7], and for Co films separated by Cu [8,9]. The dramatic sensitivity of the exchange coupling to atomic-scale film roughness that was observed for Fe/Cr/Cr [2] has verified model calculations which show that just a small amount of roughness, even a quarter of a monolayer, is enough to obscure the short-period oscillations [10].

Oscillatory coupling has not previously been observed in Fe/Ag/Fe multilayers [1,6,11]. Only FM coupling has been observed in these films with the coupling monotonically decreasing with increasing Ag thickness and vanishing after about 7 layers. This lack of AFM coupling is unexpected since oscillatory coupling is observed for the other noble metal multilayers, Fe/Cu/Fe [11] and Fe/Au/Fe [6]. Ag should also grow well on Fe because of the small lateral mismatch to the Fe lattice. In this paper, we report the first observation of oscillatory, thickness-dependent coupling through Ag in high-quality Fe/Ag/Fe sandwich structures.

Correspondence to: Dr. J. Unguris, NIST, US Department of Commerce, Gaithersburg, MD 20899, USA.

We have found that when atomically flat Ag spacer layers are grown on Fe whisker substrates, the coupling oscillates between FM and AFM with a periodicity that can be decomposed into both long- and short-period contributions. The long-period is 5.73 ± 0.05 Ag layers (1.17 ± 0.01 nm) and the short-period is 2.37 ± 0.07 Ag layers (0.485 ± 0.014 nm). The oscillations are seen to persist for Ag layers that are up to at least 50 monolayers (10.2 nm) thick. These periodicities are consistent with the 2.4 and 5.6 layer periods that are predicted by Ag Fermi surface spanning vectors [12]. This agreement supports theoretical models of the exchange coupling in which the periodicity of the oscillations are determined by the exact size and shape of the spacer layer Fermi surface.

In addition to the FM/AFM coupling, regions of magnetization along a direction lying in the surface, but perpendicular to the FM/AFM alignment, were also observed. This type of coupling was previously observed in Fe/Cr/Fe [2,13] and Co/Cu/Co [14]. The perpendicular coupling is attributed to a 'biquadratic' exchange coupling, whereas the FM/AFM alignment results from the usual 'bilinear' exchange coupling. The Fe/Ag/Fe biquadratic coupling is found to be very sensitive to the thickness of the Fe film overlayer, becoming relatively stronger as the Fe film thickness increases.

2. Experimentals

The experimental procedures used to investigate the Fe/Ag/Fe coupling were similar to those used in previous measurements of Fe/Cr/Fe [2]. The magnetization, crystalline order, and chemical composition of the films are measured using scanning electron microscopy with polarization analysis (SEMPA) [15], reflection high-energy electron diffraction (RHEED), and scanning Auger microscopy, respectively. All of the analysis is done in situ and, when necessary, during film evaporation. The films are grown on the (100) surface of a single-crystal Fe whisker substrate. The Fe whiskers are used because extremely flat, well-ordered surfaces can be obtained following well established cleaning procedures [16]. Scanning tunnelling microscope images of whiskers taken from the same growth 'boat' show a step density of only about one single atomic height step per square μm [17]. RHEED patterns from the bare whiskers show sharp diffraction spots distributed along Laue rings. The only major defects present are isolated Fe mounds, separated by 10–100 μm that are usually found near the edges of the whisker. Immediately before thin film deposition, the Fe whiskers are cleaned by Ar ion sputtering followed by annealing to 800°C. Auger spectra indicate that this cleaning procedure reduces the

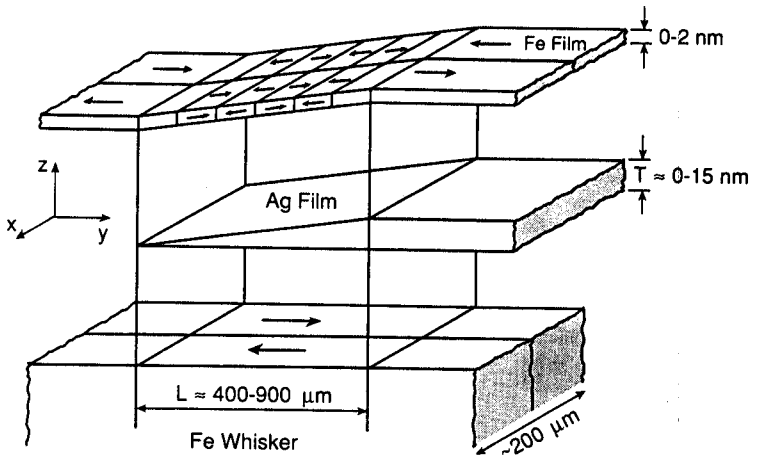

Fig. 1. An exploded schematic view of the of the sample structure showing the Fe(100) single-crystal whisker substrate, the evaporated Ag wedge, and the Fe overlayer. The arrows in the Fe show the magnetization direction in the two domains.

surface contamination, which was mostly oxygen, to below 0.05 monolayers.

After the whisker cools down from the anneal, a Ag wedge is deposited on the whisker by gradually moving a shutter in front of the sample during evaporation. A schematic drawing of the wedge is shown in fig. 1. The slope of the wedge is very small, about 0.001°. The crystalline order and thickness of the Ag wedge are measured using scanning RHEED [17]. The wedge is then coated with a thin, approximately 1 nm thick, Fe film. The type of magnetic coupling is then determined by using SEMPA to measure the direction of the magnetization in the top Fe layer. An Fe whisker that is divided lengthwise into two oppositely magnetized domains is used in order to test for and eliminate any instrumental offsets in the SEMPA measurements.

3. Ag growth results

The optimum conditions for Ag growth were determined by measuring RHEED profiles and thickness-dependent intensity oscillations from Ag wedges grown at various substrate temperatures and Ag evaporation rates. The best growths are obtained for substrate temperatures between 60 and 100°C, and for evaporation rates between 0.07 and 0.8 monolayers/s. RHEED patterns from Ag films grown at slower evaporation rates show additional diffraction spots, indicative of rougher, more three-dimensional growth. Ag films grown at higher temperatures yield two-dimensional RHEED patterns, that are as sharp as at lower temperatures, but SEMPA measurements and a lack of RHEED intensity oscillations indicate poor-quality film growth. This surprising result is understood by examining the topography of these films with the scanning electron microscope which shows that the films are broken-up into micrometer-sized regions. The size of these regions indicates that the steps in the substrate Fe whisker may possibly influence the growth at elevated temperatures. Even though the lateral mismatch between the bcc Fe and fcc Ag rotated by 45° is only 0.8%, the single atom Ag layer spacing perpendicular to the surface is 42% larger than the Fe. This mismatch may introduce significant strain at Fe steps which results in the formation of the mosaic structure at higher temperatures.

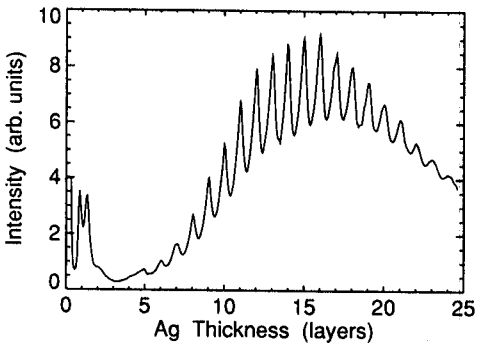

Fig. 2. The RHEED intensity as a function of Ag thickness obtained by scanning the focused incident electron beam along the uncovered Ag wedge.

The Ag growth quality as a function of Ag thickness is qualitatively evaluated by measuring the RHEED intensity along the length of the wedge. Such a RHEED scan is shown in fig. 2 for a Ag wedge grown under optimal conditions. The line scan is obtained by measuring the intensity of the specular diffraction beam as the focused incident electron beam is rastered across the specimen. For this measurement, the incident electron energy is 10 keV and the incident angle is about 2°. The line scan shows that the Ag grows in several thickness dependent phases. For the first 3–4 layers the RHEED pattern shows significant disorder and roughness, and the RHEED intensity oscillations are barely visible, aside from a transient near one monolayer. As the Ag spacer becomes thicker, the quality of the growth improves and strong RHEED oscillations are observed between 6 and 20 to 30 layers. Over this thickness range the RHEED oscillations develop a cusp-like shape which is another indication of high-quality layer-by-layer growth [19]. However, the RHEED pattern indicates that the Ag never becomes as well ordered as the bare Fe substrate. Finally, in the thicker part of the wedge, the Ag growth front becomes rougher and the oscillations become smaller and eventually disappear after about 50 layers of total thickness.

3.1. Oscillatory coupling results

Examples of SEMPA and RHEED images obtained from the same sample are shown in fig. 3. The thickness of the Ag wedge is measured using the intensity oscillations in the bare Ag scanned RHEED image. The RHEED intensity maxima are assumed to correspond to integer layer coverages. The RHEED and SEMPA images are registered by aligning defect structures common to both images. By measuring the Ag thickness with the scanning RHEED technique we avoid any errors in the thickness measurement due to time-dependent variations in the evaporation rates or due to local irregularities in the shape of the shutter. The small distortion in the thickness contours in the RHEED image is the result of such a rough spot on the shutter. After coating the Ag wedge with 3 layers of Fe the SEMPA images of the in-plane magnetization components, M_y and M_x, were obtained. M_y is the magnetization component along the magnetization direction of the Fe whisker substrate. Oscillations between FM and AFM coupling are most clearly visible in this magnetization component. Note that while this method of measuring the

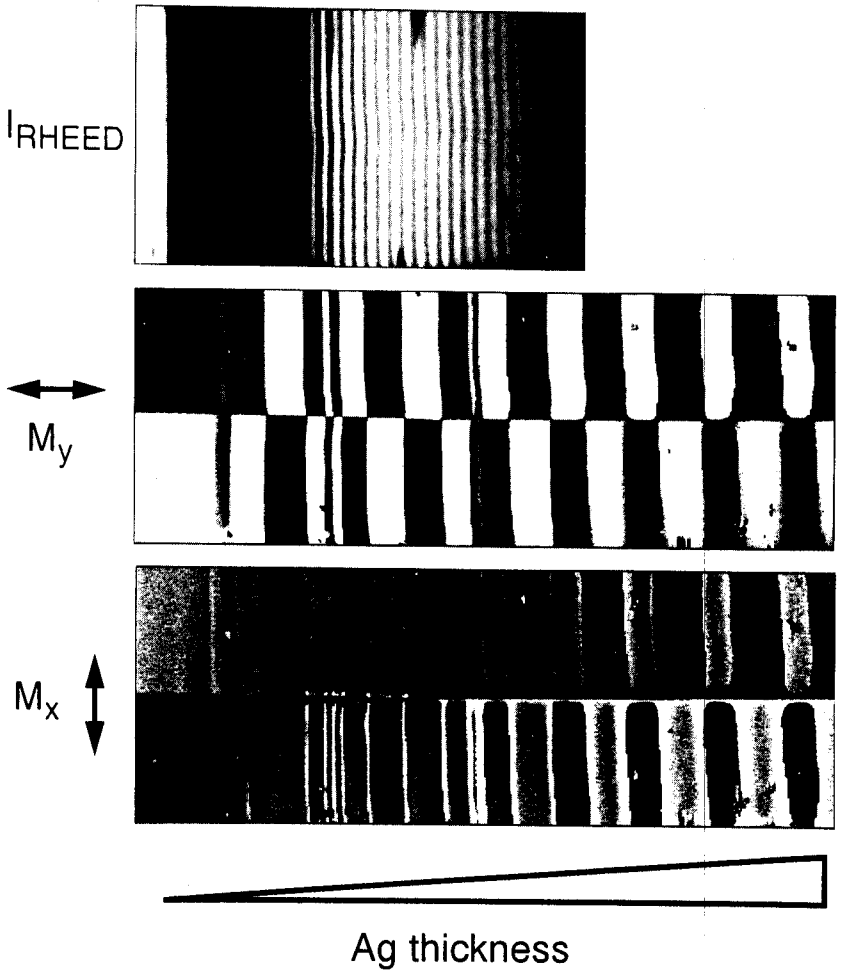

Fig. 3. A scanned RHEED image of an uncovered Ag wedge, and SEMPA images, showing the in-plane magnetization components, M_y and M_x, of the same wedge after coating it with 3 layers of Fe. The RHEED image was used to calibrate the Ag thickness.

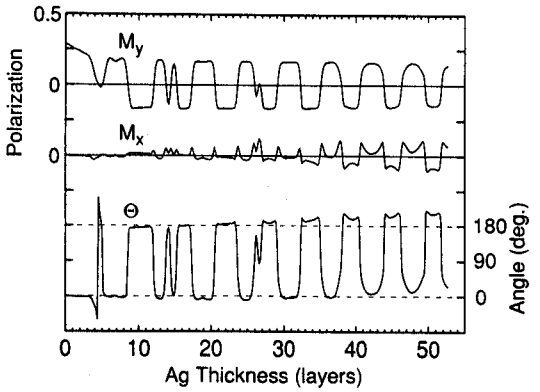

Fig. 4. The Ag thickness dependence of M_y, M_x, and the in-plane magnetization angle, θ, derived from the images in fig. 3. The angle is plotted so that 0° (180°) corresponds to FM (AFM) coupling.

coupling is extremely sensitive to whether the coupling is FM, AFM or zero, it does not give a direct measurement of the strength of the coupling. M_x is the simultaneously measured in-plane magnetization component orthogonal to the whisker axis. This component emphasizes the widths of the FM/AFM transition zones, and therefore the relative strength of the biquadratic coupling. Vector addition of these two magnetization components gives the magnitude and direction of the in-plane magnetization vector.

Fig. 4 shows M_y, M_x and the magnetization angle derived from the images in fig. 3 and plotted as a function of Ag thickness. Several noteworthy features of the Fe/Ag/Fe magnetic coupling are immediately obvious from the magnetization data:

(1) The first fluctuation in the magnetic coupling occurs at a Ag thickness of about 4 layers. In this initial oscillation the Fe film does not rotate all the way into the AFM orientation, however. The first complete AFM coupling reversal occurs after 8 layers.

(2) The coupling appears to oscillate with a long-period that varies between 5.3 and 6.4 layers. We will show that this variation results from the presence of two different oscillatory coupling periods.

(3) Distinct oscillations in the magnetic coupling with a period of about two layers are observed at Ag thicknesses of 14 and 26 layers. The appearance of these oscillations, especially the one at 26 layers, depends sensitively on the quality of the film growth. They only appear in the better-quality film growth.

(4) At least eight complete coupling oscillations are observed over a Ag thickness range of about 11 nm or 53 layers.

(5) Finally, the transition zones between FM and AFM coupling become wider as the Ag wedge becomes thicker. This broadening of the transitions suggests that the relative strength of the biquadratic coupling increases as the Ag spacer becomes thicker and/or rougher.

In order to be more quantitative about the coupling periodicities, we model the SEMPA results by using the following procedure. First, we Fourier transform the M_y data in order to get approximate values for the two major periodic components, d_1 and d_2. We then model the interlayer coupling beginning with the function,

$$f = A_1 \sin(2\pi t/d_1 + \Phi_1) + A_2 \sin(2\pi t/d_2 + \Phi_2), \quad (1)$$

where Φ_1 and Φ_2 are the adjustable phases, and A_1 and A_2 are the adjustable amplitudes of the respective periodic components. This continuous function is then discretized at the Ag lattice values so that each monolayer corresponds to only one coupling strength. The thickness at which the magnetization changes sign is determined by taking the weighted average of the coupling from adjacent monolayers. Finally, all positive values of the coupling function are set to the same positive magnetization value and all negative coupling values are set to the equal but opposite magnetization. The exact coupling periods and uncertainties are determined by varying the periodicity and searching for the best match between the calculated and measured magnetizations. For each value of the periods, the phases and amplitudes are adjusted to optimize the fit. In fig. 5 the result of the best fit model calculation is shown and compared with measurements from two different Fe/Ag/Fe wedges. For this calculation: $d_1 = 2.37$ layers, $d_2 = 5.73$ layers, $\Phi_1 = 4.80$, $\Phi_2 = -0.43$, and $A_1/A_2 = 1.02$. Except for some extra short-period oscillations at thicker coverages, the

model reproduces all of the oscillatory features in the data. The lack of short-period fine structure in the data at higher coverages is not surprising since this structure is very sensitive to roughness and our RHEED measurements show that the thicker Ag grows rougher. Including roughness in the model that is only on the order of 10% of a layer is enough to eliminate most of the small structure from the calculation.

Measurements of the thickness dependence of the magnetic coupling, and in particular, of the oscillatory coupling period(s) provide a simple test of different theoretical coupling models. In particular, for models which use an Ruderman–Kittel–Kasuya–Yoshida (RKKY)-type interaction to generate the coupling, the periodicity of the coupling is directly related to the size and shape of the Fermi surface of the spacer material. Specifically, the periodic response of the material is dominated by wavevectors that are perpendicular to the interface and span nearly parallel parts of the Fermi surface. There are two determinations of Ag(100) Fermi surface spanning vectors that are currently available and yield similar values. Bruno and Chappert [12] use a nearly free electron fit to de Haas–van Alphen and cyclotron resonance measurements of Fermi surface extremals as the basis for their Fermi surface calculations of the noble metals. For Ag(100) they predict coupling periodicities of 5.58 and 2.38 layers. Stiles [20] obtains similar periodicities using a tight-binding fit to a local density approximation Ag band structure and from that computes the interface Fermi surface spanning vectors. Thus the calculated short oscillation period is within the $\pm 3\%$ error estimate of our measured value, while the predicted long-period is about 2.5% larger than the measured period with its $\pm 1\%$ error estimate. This small descrepancy might be related to uncertainties in the calculations or to some tetragonal distortion of the Ag due to the small lattice mismatch with Fe.

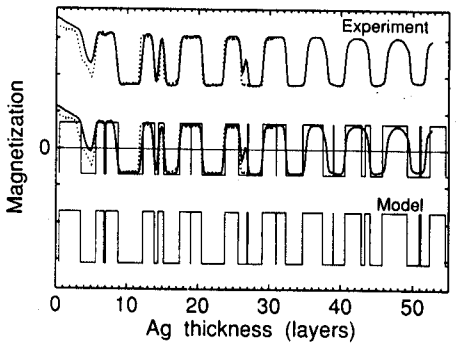

Fig. 5. The results of the model calculation described in the text are compared with the M_y magnetization measurement from fig. 4 along with a similarly processed SEMPA measurement from a different wedge (dotted line). The calculated (bottom) and measured (top) curves are shown both separately and overlaid. 2.37 and 5.73 Ag layer coupling periods were used in the model calculation.

3.2. Biquadratic coupling results

In addition to the FM/AFM coupling, the Fe/Ag/Fe sandwiches also exhibit significant biquadratic coupling. Phenomenologically, the exchange coupling energy per unit area can be written as [13]:

$$E = A_{12}[1 - \boldsymbol{m}_1 \cdot \boldsymbol{m}_2] + \tfrac{1}{2}B_{12}\left[1 - (\boldsymbol{m}_1 \cdot \boldsymbol{m}_2)^2\right], \tag{2}$$

where $\boldsymbol{m}_1$ and $\boldsymbol{m}_2$ are the unit magnetization vectors in the two Fe layers, and A_{12} and B_{12} are the bilinear and biquadratic coupling coefficients, respectively. The exchange coupling in the multilayers is usually dominated by the bilinear term, except at the transitions between FM/AFM coupling where A_{12} reverses sign and the average value of the bilinear coupling goes through zero. In these transition regions the biquadratic coupling dominates and the magnetization of the top Fe film is no longer parallel or antiparallel to the Fe whisker magnetization. The width of this transiton zone is therefore a qualitative measure of the relative strength of the biquadratic coupling. Note that these transition regions are not simply domain walls in the Fe film, since they are much wider than domain walls and they scale inversely with the slope of the Ag wedge. In order to make the relationship between B_{12} and the direction of the magnetic coupling more quantitative, the total magnetic energy of the multilayer, including crystalline anisotropy, intralayer exchange and magnetostatic contributions, must be calculated and minimized [13].

One interesting feature of the Fe/Ag/Fe coupling that was not observed in our previous Fe/Cr/Fe measurements is the sensitivity of the biquadratic coupling to the thickness of the top Fe layer. Figure 5 shows SEMPA measurements of the same Ag wedge covered with two different thickness Fe layers. M_y, M_x and a line scan of the magnetization angle are shown for a wedge coated with 3 layers of Fe and for the same wedge after an additional 4 layers were deposited (7 Fe layers in total). For both Fe coverages the relative strength of the biquadratic coupling, judging by the transition zone widths, increases with increasing Ag thickness, possibly due to a decrease in the bilinear coupling and an increase in the Ag roughness for thicker Ag interlayers. In addition as the Fe coating becomes thicker, the widths of the FM/AFM transitions become larger and in the thicker part of the Ag wedge the FM/AFM coupled regions have disappeared entirely leaving behind only 90° coupled domains. These results suggest that the relative strength of the biquadratic coupling increases as the Fe film becomes thicker. Note, however, that in addition to the change in the width of the FM/AFM transition, the shape of the transition also changes. For the thin Fe film the line scan of the magnetization angle shows that the magnetization

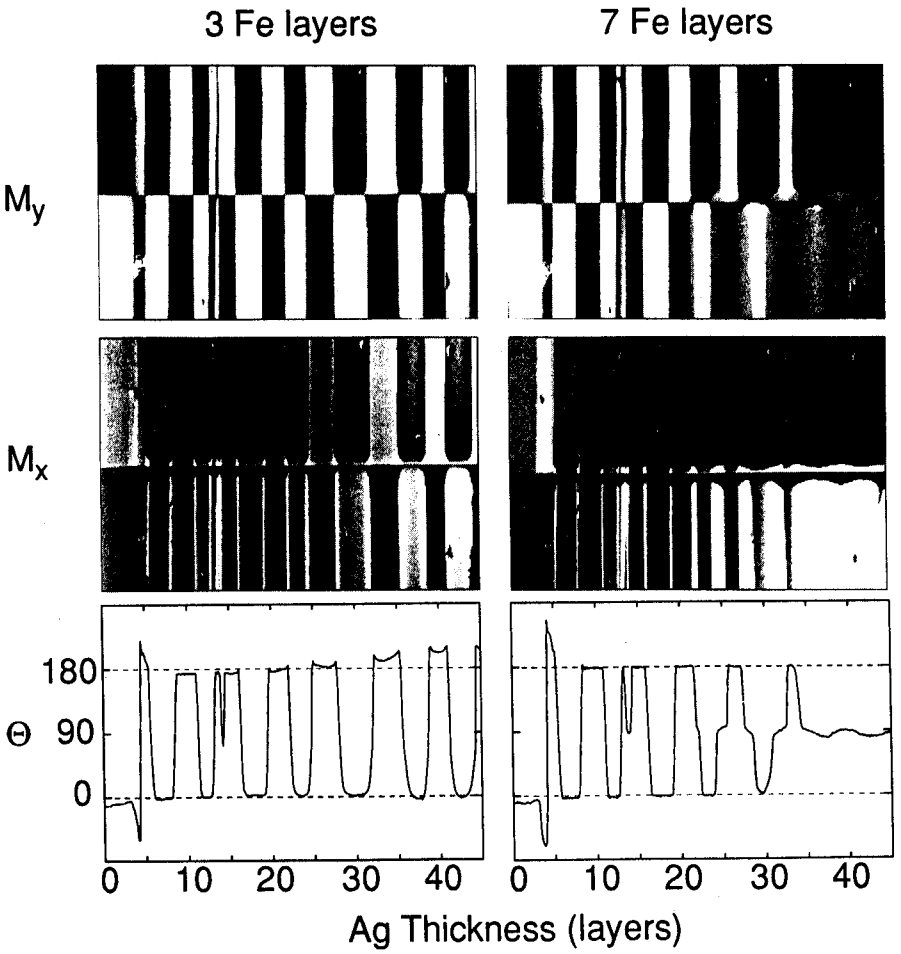

Fig. 6. Magnetization images of M_y and M_x, and a line scan of the magnetization angle, θ, of the same wedge are shown for a 3 layer Fe overlayer, and after increasing the Fe overlayer thickness to 7 layers.

rotates almost continuously across the FM/AFM transtion, while in the thicker Fe film the magnetization in the transition region prefers to be orthogonal to the whisker magnetization. This change in the transition shape is due to the increased strength of the bulk crystalline anisotropy in the thicker Fe film. In relating the relative strength of the biquadratic coupling to the FM/AFM transtion width, therefore, these changes in film anisotropy must also be taken into account.

The thickness dependence of the biquadratic coupling provides a possible test of theories regarding the as yet unresolved origin of the biquadratic coupling. The possibility that the biquadratic coupling is the result of a higher-order interaction term in the coupling Hamiltonian has been suggested [13]. In this case the biquadratic coupling does not depend on the Fe film thickness. An alternative explanation has been proposed by Slonczewski [21] in which the biquadratic coupling is the result of spatial fluctuations in the bilinear coupling caused by thickness fluctuations in the spacer layer. At coverages where the thickness fluctuations correspond to reversals in the sign of the bilinear coupling, the magnetization will favor the 90° coupling. For Fe films that are thin relative to the island size Slonczewski's model predicts a biquadratic coupling:

$$B_{12} = -2(\Delta A_{12})^2 L^2/(\pi^4 AD), \qquad (3)$$

where ΔA_{12} is the fluctuation in the bilinear interlayaer exchange coupling, L is a characteristic island length scale, A is the Fe–Fe intralayer exchange, and D is the Fe film thickness. The model therfore predicts that the strength of the biquadratic coupling should decrease with increasing Fe thickness. Our observations indicate the opposite trend, but other film parameters, such as exchange, anisotropy and roughness may also change with film thickness and these contributions to the total magnetic energy of the mulilayer should also be included in order to derive B_{12} from our magnetization measurements.

4. Conclusion

We have shown that by growing well ordered epitaxial Ag films on Fe whiskers it is possible to observe oscillatory exchange coupling in Fe/Ag/Fe(100). The coupling oscillates with a long-period of 5.73 ± 0.05 Ag layers and a short-period of 2.37 ± 0.07 Ag layers. The observed periods are consistent with RKKY-like coupling theories in which oscillation periods are derived from Fermi surface spanning vectors. In addition to the bilinear coupling, biquadratic coupling was also observed. The strength of the biquadratic coupling relative to the bilinear coupling was found to increase with increasing Fe overlayer thickness. This work emphasizes that an understanding of the exchange coupling in magnetic multilayers and the possibility of fruitful comparisons of measurements to theory depends critically on our ability to grow well characterized, nearly atomically perfect multilayers structures.

Acknowledgements

We wish to thank M.H. Kelley, M.D. Stiles and J.A. Stroscio for many helpful discussions. The Fe whiskers were grown at Simon Fraser University under an operating grant from the National Science and Engineering Research Council of Canada. This work is supported by the Office of Technology Administration of the Department of Commerce and by the Office of Naval Research.

Note

Since the submission of this paper, we have received a preprint describing optical and ferromagnetic resonance observations of coupling oscillations in Fe/Ag/Fe [22].

References

[1] S.S.P. Parkin, Phys. Rev. Lett. 67 (1991) 3598.
[2] J. Unguris, R.J. Celotta and D.T. Pierce, Phys. Rev. Lett. 67 (1991) 140.

[3] 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 (1991) 903.
[4] S. Demokritov, J.A. Wolf and P. Grünberg, Europhys. Lett. 15 (1991) 881.
[5] S.T. Purcell, M.T. Johnson, N.W.E. McGee, R. Coehoorn and W. Hoeving, Phys. Rev. B 45 (1992) 13064.
[6] A. Fuss, S. Demokritov, P. Grünberg and W. Zinn, J. Magn. Magn. Mater. 103 (1992) L221.
[7] Z.Q. Qiu, J. Pearson, A. Berger and S.D. Bader, Phys. Rev. Lett. 68 (1992) 1398.
[8] M.T. Johnson, S.T. Purcell, N.W.E. McGee, R. Coehoorn, J. aan de Stegge and W. Hoving, Phys. Rev. Lett. 68 (1992) 2688.
[9] M.T. Johnson, R. Coehoorn, J.J. de Vries, N.W.E. McGee, J. aan da Stegge and P.J.H. Bloemen, Phys. Rev. Lett. 69 (1992) 969.
[10] Y. Wang, P.M. Levy and J.L. Fry, Phys. Rev. Lett. 65 (1990) 2732.
[11] Z. Celinski and B. Heinrich, J. Magn. Magn. Mater. 99 (1991) L25.
[12] P. Bruno and C. Chappert, Phys. Rev. Lett. 67 (1991) 1602.
[13] M. Ruhrig, R. Schafer, A. Hubert, R. Mosler, J.A. Wolf, S. Demokritov and P. Grünberg, Phys. Stat. Solidi (a) 125 (1991) 635.
[14] B. Heinrich, J.F. Cochran, M. Kowalewski, J. Kirschner, Z. Celinski, A.S. Arrott and K. Myrtle, Phys. Rev. B 44 (1991) 9348.
[15] M.R. Scheinfein, J. Unguris, M.H. Kelley, D.T. Pierce and R.J. Celotta, Rev. Sci. Instrum. 61 (1990) 2501.
[16] S.T. Purcell, A.S. Arrott and B. Heinrich, J. Vac. Sci. Technol. B 6 (1988) 794.
[17] J. Stroscio, unpublished.
[18] J. Unguris, R.J. Celotta and D.T. Pierce, Phys. Rev. Lett. 69 (1992) 1125.
[19] M.G. Lagally, D.E. Savage and M.C. Tringides, in: Reflection High-Energy Electron Diffraction an Reflection Electron Imaging of Surfaces, eds. P.K. Larsen and P.J. Dobson, (Plenum, New York, 1988) p. 139.
[20] M.D. Stiles, unpublished.
[21] J.C. Slonczewski, Phys. Rev. Lett. 67 (1991) 3172.
[22] Z. Celinski, B. Heinrich and J.F. Cochran, J. Appl. Phys. to be published.