

Magnetic coupling through Cr: Study of spin polarization in Cr and film-growth effects

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The interlayer magnetic coupling of polycrystalline Fe/Cr/Fe and $\text{Ni}_{80}\text{Fe}_{20}/\text{Cr}/\text{Ni}_{80}\text{Fe}_{20}$ trilayers has been studied with spin-polarized secondary-electron emission (SPSEE) and magneto-optic Kerr effect. The exchange coupling between the two magnetic layers is observed to oscillate between ferromagnetic and antiferromagnetic as a function of Cr spacer thickness. The sign of the coupling depends on the temperature at which the trilayer structures are grown but is not affected by further changes due to contamination or interdiffusion (~ 2 atomic layers) at the interface resulting from post deposition thermal cycling up to 450 K. A $\text{Ni}_{80}\text{Fe}_{20}/(12 \text{ \AA} \text{ Cr})/\text{Ni}_{80}\text{Fe}_{20}$ structure deposited at $T=90 \text{ K}$ is antiferromagnetically coupled for $T=90\text{--}450 \text{ K}$, whereas the same structure deposited at 300 K shows a reversible transition between ferromagnetic and antiferromagnetic coupling as a function of temperature between 180 and 330 K. SPSEE measurements of Cr overlayers on both Ni-Fe and Fe set an upper limit to Cr polarization effects at 2×10^{-3} , which has important ramifications for theoretical attempts to understand the coupling mechanism.

INTRODUCTION

Magnetic interlayer coupling phenomena in layered thin-film structures have attracted considerable attention in recent years, in part because of the prospect of artificial layered structures with novel magnetic properties. One point of interest is the coupling of two ferromagnetic layers through a nonmagnetic spacer layer. In Fe/Au/Fe, for example, the ferromagnetic (FM) coupling decreases with increasing Au thickness and disappears for interlayer thicknesses larger than 20 Å but never changes sign.¹ In striking contrast, it was discovered that epitaxial Fe/Cr/Fe exhibits antiferromagnetic (AFM) coupling for certain Cr thicknesses.¹ Several groups have studied this phenomenon using different techniques and the reported Cr thicknesses x that result in AFM coupling differ somewhat: $x=4\text{--}9$,¹ 25,² 8–15,³ 9–18,⁴ 9–20,⁵ and 12–25 Å.⁶ One group observed decoupling of the two magnetic layers for $x \geq 16 \text{ \AA}$,³ whereas another group reported strongest coupling for $x=16 \text{ \AA}$.⁶ These discrepancies may be attributed to differences in film structure as well as to uncertainties in the film-thickness calibration.

More recent experiments have shown that the interlayer coupling does not just change from FM to AFM once, it oscillates as a function of spacer thickness. This oscillation has been observed in the Fe/Cr/Fe, Co/Cr/Co, and Co/Ru/Co systems⁷ as well. Superlattices of these materials which showed AFM coupling also showed the giant magnetoresistance effect.^{4,10} Most of the reported measurements are of epitaxially grown layers, but the AFM coupling does not depend on ideal single crystallinity. For example, epitaxially grown layers of Fe/Cu/Fe that exhibited AFM coupling did not show a change in coupling properties when a change in structure was observed.¹¹ Even polycrystalline films with nonideal interfaces have now exhibited AFM coupling^{7,12} in con-

trast to earlier experiments with polycrystalline $\text{Ni}_{80}\text{Fe}_{20}/\text{Cr}/\text{Ni}_{80}\text{Fe}_{20}$ which did not.¹³ This discrepancy can be attributed to differences in film structure, which is often not as well controlled in polycrystalline films.

The theoretical explanation for this coupling oscillation is currently a subject of intense debate. The decaying oscillatory behavior of the coupling is strongly suggestive of a Ruderman-Kittel-Kasuya-Yosida (RKKY)-type coupling but the wavelength of the oscillations is unexpectedly large (15–20 Å). A number of different theoretical approaches have been investigated,^{1,14–19} but at this point there is no generally accepted model that will explain the experimental observations. A few general comments can be made about this phenomenon. First, the antiferromagnetic properties of bulk Cr cannot be involved because the AFM coupling exists at temperatures higher than the Néel temperature of Cr. In addition, AFM coupling has been observed with spacers of Cu and Ru as well. Second, calculations prove that the coupling is much too strong to be of dipolar origin.²⁰ Further experimental work is needed to explore the influence of film structure and interface quality on the strength and thickness dependence of the coupling.

Two different experimental approaches have been employed to study this interlayer coupling. The first is to deposit a multilayer and measure the interlayer coupling by *ex situ* magnetometry in which the magnetization curve or the ferromagnetic resonance of the whole stack is used to determine the strength of the coupling. The second is to deposit a trilayer structure and use surface sensitive techniques. In the present work the second approach was used to study the interlayer coupling through Cr *in situ*, primarily by spin-polarized secondary-electron emission (SPSEE). In contrast to the *ex situ* techniques, the SPSEE magnetic signal comes from the top surface of a multilayer rather than from the entire structure. This study is therefore very closely related to the early experi-

ments on epitaxial Fe/Cr structures by Carbone *et al.*² The recently discovered oscillatory behavior of the Cr (Ref. 7) coupling motivated us to reexamine this phenomenon with SPSEE. If a RKKY mechanism is producing the coupling, the Cr conduction electrons should be spin polarized and the polarization should display oscillations that correspond to the oscillations of the interlayer coupling. A measurement of the Cr polarization is therefore an important part of the investigation of this coupling mechanism.

The present results show that ion beam deposited polycrystalline Fe/Cr/Fe and $\text{Ni}_{80}\text{Fe}_{20}/\text{Cr}/\text{Ni}_{80}\text{Fe}_{20}$ trilayers exhibit oscillatory coupling behavior. No statistically significant Cr magnetization was seen in the present study, establishing an upper limit of 2×10^{-3} for the Cr spin polarization. The effects on the coupling of deposition temperature, post deposition thermal cycling, and interface contamination will also be discussed.

EXPERIMENT

The apparatus used in this experiment has been described in detail elsewhere.²¹ Magnetic information was obtained by magneto-optic Kerr effect (MOKE) and SPSEE. The low-energy cascade electrons were excited by a primary unpolarized 3.2-keV electron beam and were imaged with an extraction lens system onto the entrance diaphragm of a medium energy (40-keV) Mott spin-polarization analyzer.²¹

The probing depth for MOKE is $\sim 100 \text{ \AA}$, whereas the probing depth of SPSEE is $\sim 5 \text{ \AA}$ (for transition metals).²²⁻²⁴ Figure 1 illustrates the complementary nature of these techniques which result from their different probing depths. Hatched areas indicate ferromagnetic, open areas nonmagnetic material, and the probing depth of the measurement is indicated by shading. (1) represents the surface of a ferromagnet, (2) an ultrathin ferromagnetic film on top of a nonmagnetic substrate, and (3) an ultrathin magnetic overlayer separated from a ferromagnetic substrate by a nonmagnetic spacer. In all three cases the left-hand side labeled (a) schematically shows the probing depth of MOKE and the right-hand side labeled (b) shows the probing depth of SPSEE. In (1), (b) may reveal properties of the very surface inaccessible to (a). In (2), the magnetic information obtained with both techniques is expected to be equivalent. (3) is the most interesting case because here (a) and (b) are complementary measurements. If the magnetic substrate is much thicker than the magnetic overlayer, (a) will mainly see the magnetic substrate and (b) will mainly see the magnetic overlayer, so comparison of hysteresis loops from MOKE and SPSEE will yield the relative magnetic alignment of the two layers.

The films were sputter deposited onto a 4-mm wide polished Cu band by Xe ions from a plasma gun directed at a sputtering target. The Cu band was mounted on a ultrahigh vacuum (UHV) precision manipulator which could be cooled to 90 K or heated to 450 K. An electric current flowing through the band was used to produce an in-plane homogeneous magnetic field of up to 25 Oe.

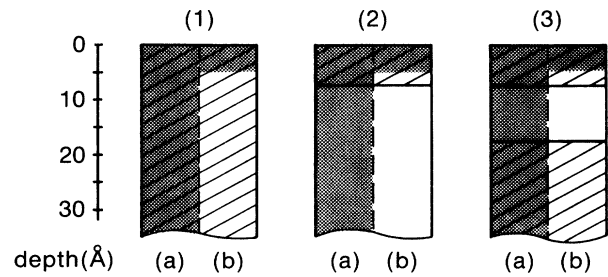


FIG. 1. Schematic diagram of probing depth (shaded area) of (a) MOKE and (b) SPSEE. The hatched area is ferromagnetic and the open area nonferromagnetic. (1) Represents the surface of a ferromagnet, (2) an ultrathin ferromagnetic film on top of a nonmagnetic substrate, and (3) an ultrathin magnetic overlayer separated from a ferromagnetic substrate by a nonmagnetic spacer.

Both MOKE and SPSEE measurements were configured to measure magnetization components along the applied magnetic field. Film thicknesses were measured by a previously calibrated quartz microbalance. The absolute thickness error is estimated to be $\pm 15\%$, but the relative error is less than $\pm 5\%$. The chemical composition of the films as well as the growth behavior was monitored by Auger electron spectra (AES) obtained with a single pass cylindrical mirror analyzer.

To make the trilayer structures described here, first several hundred Å thick permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) film was deposited at room temperature in the presence of a magnetic field. In this way Ni-Fe films with uniaxial anisotropy were obtained leading to square loops with coercivity $H \leq 10 \text{ Oe}$ as verified by MOKE and SPSEE. Square loops with low coercivity were essential because the low-energy secondary cascade electrons were extracted in a direction perpendicular to the applied magnetic field. Measurement of complete hysteresis loops is essential to insure that the sample is in a single domain state. Thin Fe layers deposited on top of the Ni-Fe film were found to be strongly coupled to it and exhibited the same hysteresis behavior, although with increased spin polarization due to the larger magnetic moment in Fe.²⁵ In this way it is possible to produce a low coercivity polycrystalline Fe surface by overwhelming the intrinsic anisotropies by strong exchange coupling to the thicker Ni-Fe film. Each magnetic thin-film structure was deposited onto an underlayer of several hundred Å of Cr which provided magnetic isolation from the underlying layers.

Figure 2 shows an example of SPSEE hysteresis-loop measurements of the film structure $(400\text{-Å Ni-Fe})/(10\text{-Å Fe})/(x/\text{Cr})$. The flat wings of the loops demonstrate the single magnetic domain state of the sample, and the average of the measured spin-polarization values at magnetic saturation is taken as the value of the secondary cascade polarization P_C . The rapid polarization decrease as a function of Cr thickness proves that the short probing depth of this technique previously observed for Fe (Refs. 22 and 24) and Ta (Ref. 23) also applies to Cr.

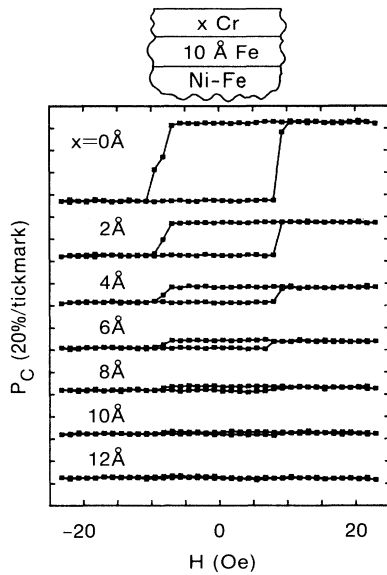


FIG. 2. SPSEE hysteresis loops of a (400-Å Ni-Fe)/(10-Å Fe) surface before and during deposition of a sequence of 2-Å thick Cr layers. x is the total Cr thickness.

THICKNESS-DEPENDENT OSCILLATIONS OF THE EXCHANGE COUPLING

Trilayer structures of two different types were studied: Fe/Cr/Fe and Ni₈₀Fe₂₀/Cr/Ni₈₀Fe₂₀. The lower Fe film in the Fe/Cr/Fe structures was deposited onto a Ni-Fe film for the reasons described above. The thickness of the top ferromagnetic layer varied from 5 to 15 Å, but this variation did not affect the sign of the coupling. Except for a few special cases, which will be described in another section of this paper, the Cr spacer layers and the top ferromagnetic layers were deposited at $T=90$ K. The lower of the two ferromagnetic layers was purposely made much thicker than the top layer so it would line up with the applied magnetic field. In this case, the sign of the SPSEE hysteresis loop directly determines the parallel or antiparallel alignment of the layers without further comparison with the MOKE loop.

It may seem straightforward to assume that observed parallel and antiparallel configurations are necessarily caused by FM and AFM coupling across the Cr spacer layer, but it is important to discuss this relationship in more detail. A parallel alignment could be due to FM coupling, but could also be produced by the absence of coupling. Since we measure the full SPSEE loop, not just the remanent polarization, we have another important measure of coupling, the coercivity of the top layer. Total decoupling should cause a marked difference between the coercivity of the underlayer and the overlayer, and this coercivity difference was observed for Cr spacer layers with a thickness larger than 50 Å. This observation indicates that stray field coupling (e.g., domain-wall coupling) is most likely not what is keeping the two coercivities equal because its range would be larger than 50 Å.

Therefore, the observation of parallel arrangement of the two layers combined with the coercivity measurements proves the existence of genuine ferromagnetic coupling. Antiparallel alignment of the layers can only be due to an effective AFM coupling, either of dipolar or exchange origin. The dipolar origin for our observed AFM coupling was ruled out by the following test. Several trilayers that showed AFM coupling were extended to five layer structures by deposition of another identical spacer and an identical thin magnetic overlayer. The combined thickness of the two overlayers was still much smaller than the underlayer thickness. If the couplings in the stack were dipole dominated, the two thin overlayers would arrange themselves parallel to one another and antiparallel to the underlayer in order to minimize the stray magnetic field. It was observed instead that the two magnetic overlayers were antiparallel, proving that exchange coupling through Cr was the dominant coupling. It is for these reasons that we can conclude that parallel-antiparallel alignment of the two magnetic layers corresponds to FM-AFM coupling through the Cr spacer.

In Fe/Cr/Fe, with a Cr spacer thickness x , we observed FM coupling for $x=4$ and 8 Å, AFM coupling for $x=12$ and 18 Å, and FM coupling again for $x=25$ Å. For 25-Å Cr, the two Fe layers were almost decoupled, but by adding 3 Å of Ni-Fe on top of the Fe overlayer, we were able to detect the FM coupling. This is in good agreement with the broad range of values reported in the literature.²⁻⁷ In Ni-Fe/Cr/Ni-Fe trilayers, we found FM coupling for $x=1-6$ Å, mixed or weak coupling for $x=7$ Å, AFM coupling for $x=10, 12,$ and 15 Å, mixed or weak coupling for $x=16$ Å, FM coupling for $x=18, 20,$ and 25 Å, AFM coupling for $x=30$ Å, and FM coupling

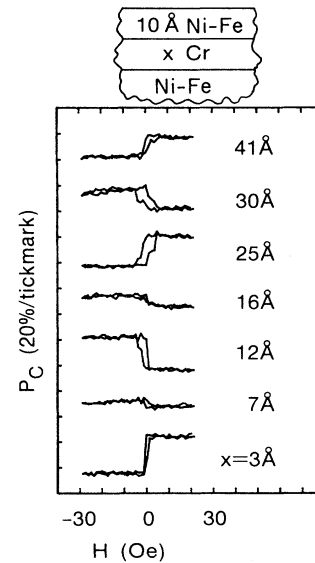


FIG. 3. SPSEE hysteresis loops of a Ni-Fe/(x Cr)/Ni-Fe trilayer structure for different values of x . The spin polarization of the secondary cascade electrons P_C is plotted vs magnetic field. The changes of sign of the hysteresis loops correspond to the oscillation of the interlayer coupling as a function of x .

for $x=41 \text{ \AA}$. For $x=50 \text{ \AA}$, the system became decoupled, as indicated by the H_C of the overlayer being different from the H_C of the underlayer. The boundaries between the FM and the AFM thickness regions showed a flat polarization signal which could be caused by a mixture of FM and AFM coupled domains or by coupling which was too weak to magnetically switch the top layer.

Figure 3 shows representative SPSEE hysteresis loops of Ni-Fe/Cr/Ni-Fe trilayers for different Cr thicknesses x . The periodic reversal of the hysteresis loops is evident. The slight differences in coercivity between the various trilayers is not significant; the coercivity of the underlayer shows the same variability.

POLARIZATION OF SECONDARY ELECTRONS FROM Cr ON Fe AND Ni-Fe

The unique capabilities of SPSEE were used to determine if these striking oscillations of interlayer coupling were accompanied by corresponding magnetization of the Cr spacer layer. The spin-density-wave value of the Cr moment is $0.6\mu_B$, which translates to an average polarization of the six Cr conduction electrons of 10%. Obviously one would not expect a detectable SPSEE signal if the Cr just orders in its bulk antiferromagnetic state. Even if the Cr layers grew with uncompensated (ferromagnetic) planes at the top surface, roughness and the finite probing depth (2–3 atomic layers) of SPSEE would suffice to wash out any observation of such localized polarization. The presence of an indirect exchange coupling through Cr strongly suggests, however, that Cr conduction electrons may acquire an additional polarization due to the contact with a ferromagnet. If such a polarization is responsible for the oscillating coupling, an oscillating polarity would be expected for the polarization as well. SPSEE is well suited to measure even small induced magnetism in Cr because its resolution, taking into account systematic errors, is about 0.1–0.2%. If one assumes that the polarization of secondary electrons from Cr is at least equal to the polarization of the Cr conduction electrons (reasonable within a factor of 2), SPSEE is therefore sensitive to an induced moment as small as $0.01\mu_B/\text{Cr-atom}$. For comparison, in (bulk) Gd, where the exchange coupling is also indirectly transmitted through the conduction electrons, the latter acquire an induced moment of $0.55\mu_B/\text{atom}$. Another important capability of SPSEE is that its probing depth is three to four times smaller than the period of the coupling oscillations. The experimental task of measuring Cr polarization by means of SPSEE is nevertheless complicated by the fact that the detected signal is not element specific. A thin layer of Cr must be grown on a ferromagnetic underlayer, and the presence of Cr polarization must be detected on top of a large polarization background emitted from the underlayer. The Cr polarization should appear as a periodic modulation, however. It should also help that the attenuation of the underlayer polarization with Cr thickness is more rapid than the expected decay of the Cr polarization (as assumed from the decay of the coupling strength⁷). The earlier spin-polarized low-energy electron diffraction (SPLEED) experiments (Carbone

*et al.*²) already indicate that Cr polarization effects, if present, are very small.

Figures 4(a) and 5(a) show the polarization of secondary cascade electrons as a function of Cr thickness for Fe/Cr and $\text{Ni}_{80}\text{Fe}_{20}/\text{Cr}$. A representative SPSEE hysteresis loop is shown on both figures. In the case of Cr on Fe (Fig. 4), a full loop was measured on the fresh Fe surface. Since the loop was essentially square, in the subsequent steps of Cr depositions, only the remanent polarization was measured. This was done to minimize any potential artifacts due to the application of an external magnetic field. In the Cr on Ni-Fe case, a full hysteresis loop was measured for each Cr thickness and the plotted polarization value is the average of all the points at magnetic saturation. In both cases, enough electrons were counted that the statistical uncertainty in the polarization would be smaller than the residual systematic errors. The combined errors limit the accuracy to an estimated $\Delta P \leq \pm 0.2\%$. Both measurements were performed at 90 K. In order to emphasize any modulation in the polarization curve, the experimental data is fitted to a simple exponential $P(x) = P_0 e^{(-x/l)}$. This would accurately describe the measured curve if the Cr secondaries were unpolarized and if the production of secondary electrons was material independent. In both cases, the fitted curve [solid lines in Figs. 4(a) and 5(a)] closely follows the measured data. The low values at zero Cr thickness may simply indicate that the surface magnetization of the ferromagnet is slightly reduced upon initial coverage with Cr. What is more important is that the values obtained

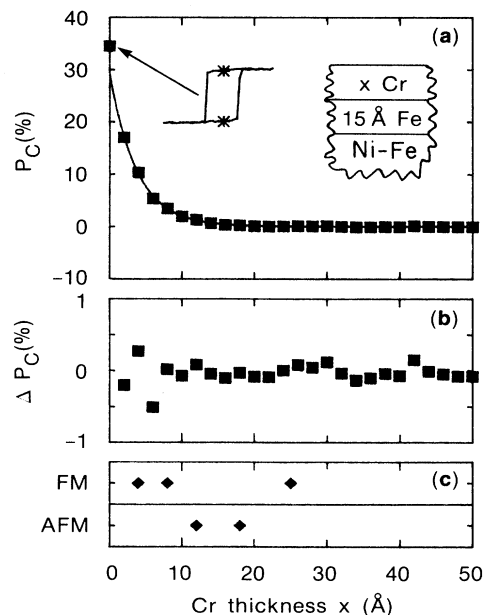


FIG. 4. (a) Remnant P_C of Cr on Fe as a function of Cr thickness x . The inset MOKE hysteresis loop verifies the loop squareness needed for remnant measurements. (b) ΔP_C , the deviation from the fitted exponential decrease as a function of x . (c) Sign of interlayer coupling, FM or AFM, in Fe/Cr/Fe trilayer structures as a function of x . There is no evidence of oscillating Cr magnetization.

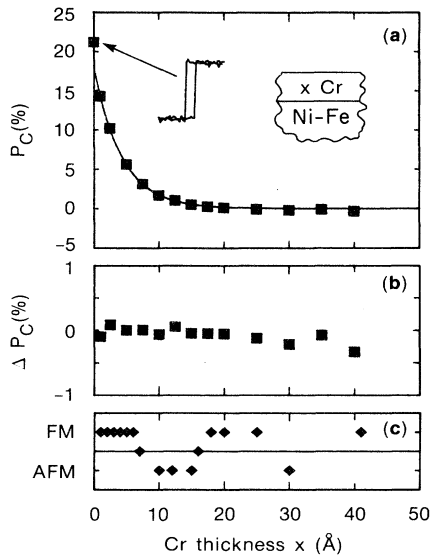


FIG. 5. (a) P_C of Cr on $\text{Ni}_{80}\text{Fe}_{20}$ as a function of the Cr thickness x . A typical SPSEE hysteresis loop is inset; the average of the measured polarization values at magnetic saturation yields P_C . (b) ΔP_C , the deviation from the fitted exponential decrease as a function of x . (c) Sign of interlayer coupling, FM or AFM, of Ni-Fe/Cr/Ni-Fe trilayer structures as a function of x . There is no evidence of oscillating Cr magnetization.

for $\lambda = 3.7 \pm 0.5$ Å for Cr on Fe and 4.3 ± 0.5 Å for Cr on $\text{Ni}_{80}\text{Fe}_{20}$ are consistent with the attenuation length expected for an unpolarized transition-metal overlayer, so there is no evidence for a monotonically decaying polarization in Cr. The bottom panels of Figs. 4(c) and 5(c) summarize the sign of the coupling observed in the trilayer experiments. The most favorable range for observation of an oscillating Cr polarization is between 10 and 20 Å, the location of the first AFM region. The strongest polarization feature (either maximum or minimum) should be located here, and the polarization background is already much reduced at this point. Expanded views of the data after background subtraction are shown in Figs. 4(b) and 5(b). In both cases, even for the first and strongest AFM region, there is no extremum in the polarization. This negative result certainly does not imply the total absence of Cr polarization. Attempts at explaining the oscillatory coupling should, however, be consistent with polarization effects in Cr no greater than 0.002 or induced moments no greater than $\sim 0.01\mu_B$.

To complete the description of the polarization versus thickness in these coupled structures, Fig. 6 shows the polarization measured during the growth of an Fe layer on Cr. The squares of Fig. 6 show, again for reference, the polarization versus Cr thickness for Cr grown on Fe. The asterisks are from a separate experiment in which the Cr deposition was interrupted at 12 Å, i.e., in the AFM coupling range, and an Fe layer was grown on the Cr. The experiment was carried out at $T = 90$ K. The AFM coupling is indicated by the negative polarization

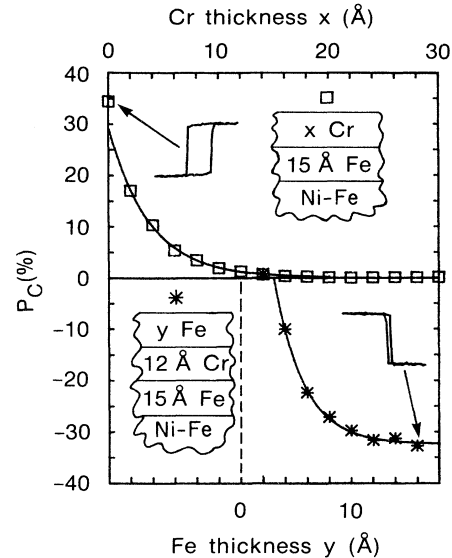


FIG. 6. P_C for Cr on Fe as a function of Cr thickness x (squares). P_C for Fe AFM coupled to Fe through a 12-Å Cr spacer as function of Fe thickness y (asterisks). A typical SPSEE loop is shown for each case.

values in the Fe layer. Also evident is a delayed onset of polarization. About 3 Å of Fe are needed for the onset of the magnetic signal, because, for 2-Å Fe, the signal is still positive and fits the exponential decrease. At about 3 Å a steep onset occurs and the data points fit an exponential increase with a characteristic length of about 3 Å. The lack of polarization in the initial Fe layer could be due to an interfacial layer with reduced magnetization, a Curie temperature below the bulk value, or perhaps that the overlayer is below a minimum thickness required for coupling.

INFLUENCE OF INTERFACIAL CHANGES ON THE COUPLING

The present results as well as the work of other groups^{7,12} show that an ideal single-crystalline interface is not required for the existence of oscillating coupling. In fact, even the local magnetic variations resulting from the distribution of Fe and Ni atoms in the alloy do not disturb the coupling. The chemical composition of the surfaces was monitored with AES and contamination by C and O was always seen. As expected, the contamination levels decreased as the experiment progressed. In the absence of depth-resolved chemical information, it is only possible to estimate that the levels of C and O were in the 10–1% range. No correlation was observed between the presence of C and O in the AES and the sign or periodicity of the coupling.

SPSEE measurements of ultrathin ferromagnetic layers are an exquisitely sensitive test for interface diffusion because dilution of the magnetic material with nonmagnetic atoms dramatically reduces the spin polarization of the secondary cascade electrons. Figure 7(a) shows P_C of a

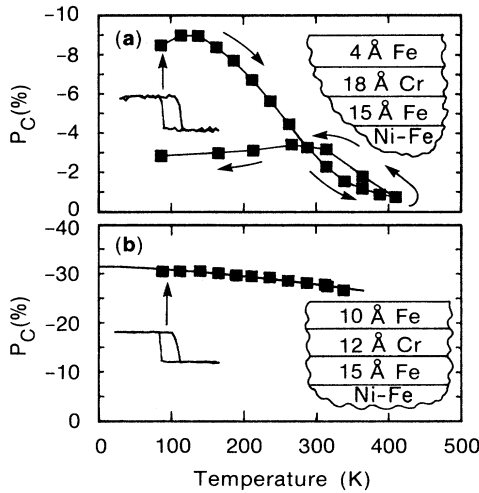


FIG. 7. (a) Temperature dependence of P_C from a 4-Å Fe film AFM coupled to Fe through an 18-Å Cr spacer. Arrows indicate the temporal order of the data points. (b) Temperature dependence of P_C from a 10-Å Fe film AFM coupled to Fe through a 12-Å Cr spacer. The temperature dependence is reversible. A typical SPSEE hysteresis loop is inset in (a) and (b).

4-Å Fe film deposited on an 18-Å Cr spacer. A representative hysteresis loop is shown and each data point is the average value at magnetic saturation. The films were deposited at $T=90$ K, warmed, and then cooled again. P_C does not come back to the same value after the heating cycle, which is evidence for interfacial diffusion. The sign of the coupling did not change, however. Figure 7(b) shows the temperature dependence of a 10-Å Fe film deposited on a 12-Å Cr spacer. The temperature dependence of this film is reversible and corresponds to the Bloch law as expected for a ferromagnetic thin film of this thickness. Since this 10-Å film shows a high value of P_C and the expected reversible temperature dependence, diffusion at the Fe/Cr interface is apparently limited to, at most, one or two layers at these temperatures.

INFLUENCE OF GROWTH TEMPERATURE ON THE COUPLING

Figure 8 shows a temperature-dependent series of SPSEE hysteresis loops of a $\text{Ni}_{80}\text{Fe}_{20}$ (12-Å Cr)(15-Å $\text{Ni}_{80}\text{Fe}_{20}$) trilayer structure. The loops show a temperature-dependent change from FM to AFM interlayer coupling. This transition was found to be reversible and reproducible, and was seen only if the Cr layer was deposited at room temperature. If the Cr layer was deposited at $T=90$ K, the coupling was AFM over the range $T=90$ –450 K. The temperature at which the Ni-Fe layers were grown made no difference. The shape of the loop at 264 K shows that the AFM coupling, which is apparent at zero external magnetic field, is so weak that an external magnetic field of ~ 20 Oe is strong enough to rotate the top magnetic layer into FM alignment. The AFM coupling obviously becomes

progressively stronger as the sample is cooled. A careful study of the film-growth mode was made with the Auger electron intensities of overlayer and substrate and their ratio following the model of Ossicini *et al.*²⁶ For all materials and growth temperatures discussed in this paper, the Auger intensity ratios showed a positive second derivative, indicating, or at least consistent with, continuous layer growth. In particular, no indication for a different growth mode was seen between the layered structures that showed the reversible FM-AFM transition and those that did not. The reversibility of the FM-AFM transition shows that it cannot be due to interdiffusion caused by heating during the measurement cycle. A plausible explanation for this striking transition is competition between two different coupling mechanisms with different temperature dependences. The two layers are AFM coupled through the Cr interface, but would be FM coupled through any pinholes that may exist in the Cr film. Since coupling through pinholes would be much stronger than coupling through the Cr spacer, only a few small pinholes are needed to produce a competitive coupling. If the coupling through the Cr spacer is weaker at high temperature, this could allow the pinhole coupling to dominate. There is no direct evidence for this explanation, but there certainly could be a few small pinholes whose number or size depended on growth temperature, and the temperature dependence of

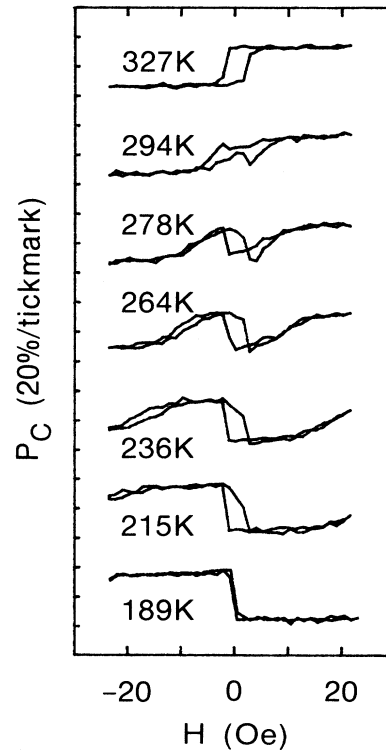


FIG. 8. SPSEE hysteresis loops from a Ni-Fe/(12-Å Cr)/(15-Å Ni-Fe) trilayer structure grown at room temperature. The magnetic interlayer coupling exhibits a reversible transition from FM to AFM when cooled.

the AFM coupling is not known. If the AFM coupling was weaker at higher temperatures, the net coupling could reproduce the effect seen here. The AES analysis of the film-growth mode is much too insensitive to detect pinholes of such small coverage.

CONCLUSION

Fe/Cr/Fe and Ni₈₀Fe₂₀/Cr/Ni₈₀Fe₂₀ polycrystalline trilayers deposited by ion beam sputtering have been investigated *in situ* with SPSEE and MOKE. The sign of the polarization in the ferromagnetic layers was used to determine their parallel or antiparallel arrangement. Full hysteresis loops, the coercivity in particular, were used to prove the existence of genuine FM and AFM exchange coupling through the Cr spacer layer. It was found that, in both systems, the coupling oscillate between FM and AFM as a function of Cr layer thickness. The wavelength is of the order of 20 Å for Fe and 16 Å for Ni₈₀Fe₂₀. The surface-sensitive nature of SPSEE provides a unique opportunity to study any induced polarization in the Cr spacer layer. SPSEE can detect magnetism present in Cr with a sensitivity of about 0.01 μB/atom integrated over a probing depth of about 4 Å. In spite of this considerable sensitivity, no evidence was found for any induced magnetism in ultrathin Cr layers deposited on Fe and NiFe. The similarity of the coupling through Cr with RKKY indirect coupling phenomena suggests that the coupling could be transmitted through polarization of the Cr conduction electrons, and it further suggests that an oscillatory polarization in Cr should be at the origin of the observed coupling oscillations. A care-

ful analysis of our polarization data reveals no oscillatory polarization component, within the accuracy of our measurement. Polarization effects associated with the first (and strongest) antiferromagnetic Cr thickness range are less than 0.002. Any future theoretical attempts to explain the coupling should be compatible with this low value.

Experimental study of the effects of temperature revealed that low deposition temperature seems to lead to more continuous Cr spacer layer growth. This was evidenced by the fact that, for a Cr thickness in the first antiferromagnetic coupling range, structures deposited at low temperature (90 K) were fully AFM coupled, whereas similar structures deposited at room temperature were AFM coupled at low temperatures but ferromagnetic above 330 K. In the latter case, the coupling through a portion of the Cr must be ferromagnetic, suggesting discontinuities in the Cr layers. It can also be inferred that the AFM coupling may decay with temperature more rapidly than the direct (pinhole) ferromagnetic coupling. A further important observation is that structures deposited at low temperature and subjected to a temperature cycle to moderately high temperatures (450 K) do interdiffuse. The interdiffusion is clearly revealed by SPSEE, from which one can also estimate that the thickness of the interdiffused layer at the interface is about 2 atomic layers.

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¹P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, *Phys. Rev. Lett.* **57**, 2442 (1986); P. Grünberg, R. Schreiber, Y. Pang, U. Walz, M. B. Brodsky, and H. Sowers, *J. Appl. Phys.* **61**, 3750 (1987).

²C. Carbone and S. F. Alvarado, *Phys. Rev. B* **36**, 2433 (1987).

³F. Saurenbach, U. Walz, L. Hinchey, P. Grünberg, and W. Zinn, *J. Appl. Phys.* **63**, 3473 (1988).

⁴M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friedrich, and J. Chazelas, *Phys. Rev. Lett.* **61**, 2472 (1988).

⁵A. Barthélémy, A. Fert, M. N. Baibich, S. Hadjoudj, F. Petroff, P. Etienne, R. Cabanel, S. Lequien, F. Nguyen Van Dau, and G. Creuzet, *J. Appl. Phys.* **67**, 5908 (1990).

⁶J. J. Krebs, P. Lubitz, A. Chaiken, and G. A. Prinz, *J. Appl. Phys.* **67**, 5920 (1990).

⁷S. S. P. Parkin, N. More, and K. P. Roche, *Phys. Rev. Lett.* **64**, 2304 (1990).

⁸D. Pescia, D. Kerkmann, F. Schumann, and W. Gudat, *Z. Phys. B* **78**, 475 (1990).

⁹S. S. P. Parkin, R. Bhadra, and K. Roche, *Phys. Rev. Lett.* **66**, 2152 (1991); J. J. de Miguel, A. Cebollada, J. M. Gallego, R.

Miranda, C. M. Schneider, P. Schuster, and J. Kirschner, in *Proceedings of the European Materials Research Society Spring Meeting, 1990, Strassbourg, France* [J. Magn. Magn. Mater. (to be published)].

¹⁰G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, *Phys. Rev. B* **39**, 4828 (1989).

¹¹B. Heinrich, Z. Celinski, J. F. Cochran, W. B. Muir, J. Rudd, Q. M. Zhong, A. S. Arrott, K. Myrtle, and J. Kirschner, *Phys. Rev. Lett.* **64**, 673 (1990); J. F. Cochran, J. Rudd, W. B. Muir, B. Heinrich, and Z. Celinski, *Phys. Rev. B* **42**, 508 (1990).

¹²M. B. Stearns, Y. Cheng, and C. H. Lee, *J. Appl. Phys.* **67**, 5925 (1990).

¹³H. Bosse and H. Gärtner, *J. Magn. Magn. Mater.* **80**, 339 (1989).

¹⁴H. Hasegawa, *Phys. Rev. B* **42**, 2368 (1990).

¹⁵Y. Wang, P. M. Levy, and J. L. Fry, *Phys. Rev. Lett.* **65**, 2732 (1990).

¹⁶W. Baltensperger and J. S. Helman, *Appl. Phys. Lett.* **57**, 2954 (1990).

¹⁷D. M. Edwards and J. Mathon (unpublished).

¹⁸F. Herman, J. Sticht, and M. Van Schlifgaarde (unpublished).

¹⁹A. Vega, A. Rubio, L. C. Balbas, J. Dorantes-Davila, S. Bouarab, C. Demangeat, A. Mokrani, and H. Dreyssé (unpublished).

²⁰P. M. Levy, K. Ounadjela, S. Zhang, Y. Wang, C. B. Som-

- mers, and A. Fert, *J. Appl. Phys.* **67**, 5914 (1990).
- ²¹D. Mauri, D. Scholl, H. C. Siegmann, and E. Kay, *Phys. Rev. Lett.* **62**, 1900 (1989); **64**, 2101 (1990); *Appl. Phys. A* **49**, 439 (1989).
- ²²M. Stampanoni, *Appl. Phys. A* **49**, 449 (1989).
- ²³M. Donath, D. Scholl, H. C. Siegmann, and E. Kay, *Appl. Phys. A* **52**, 206 (1991).
- ²⁴O. Paul, S. Toscano, and M. Landolt (unpublished); O. Paul, *Diss. Nr. 9210* Swiss Federal Institute of Technology, Zürich (1990).
- ²⁵M. Donath, D. Scholl, H. C. Siegmann, and E. Kay, *Phys. Rev. B* **43**, 3164 (1991).
- ²⁶S. Ossicini, R. Memeo, and F. Ciccacci, *J. Vac. Sci. Technol. A* **3**, 387 (1985).