# Low-temperature growth of giant magnetoresistance spin valves

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We have investigated the dependence of the giant magnetoresistance (GMR) effect, the coercivity, the coupling field, and the resistivity on film deposition at low-substrate temperatures (150 K) in spin valve multilayers of the general type: $FeMn/Ni_{80}Fe_{20}/Co/Cu/Co/Ni_{80}Fe_{20}$/glass. Low substrate temperatures tend to suppress both thermally activated surface diffusion of deposited atoms and interdiffusion at interfaces, which often occur during thin-film deposition at room temperature. We find significant increases in the GMR, significant reductions in the magnetic coupling across the Cu layer, slight reductions in the coercivity of the unpinned film, and slight reductions in the resistivity depending on which parts of the multilayer are deposited at low temperature. When the entire film is deposited at 150 K we obtain a GMR of 8.8% at a coercivity of less than 0.5 mT (5 Oe). © *1996 American Institute of Physics.* [S0021-8979(96)04401-7]

## I. INTRODUCTION

In the few years since the giant magnetoresistance (GMR) effect was discovered,[1–3] much research has been directed at attempts to increase the size of the GMR effect and to decrease the size of the magnetic field required to produce the effect. Technological applications of great economic importance are likely to result from such efforts to achieve large GMR at low fields. In the recent literature, GMR values as large as 80% at room temperature (RT) have been reported in Co/Cu superlattices[4] and saturation fields as low as 0.2 mT (2 Oe) have been reported for GMR spin valves,[5] but nothing even close to 80% GMR at 0.2 mT has even been found. Instead, the 80% GMR sample had a saturation field of ~1 T, and the sample with a coercivity of 0.2 mT had a GMR of only 3%. However, there does not appear to be any fundamental barrier, in the physics of the problem, preventing the largest values of GMR at extremely low saturation fields. If samples could be tailor-made at the atomic level with atomic perfection it should be possible to eliminate the sources of the large saturation fields in samples which exhibit large GMR values. Atomic-scale engineering of the arrangement of atoms should make it possible to reduce the coercivity, the anisotropy, and the magnetostatic coupling to almost arbitrarily low levels, and it should be possible to arrange a cancellation of the oscillatory exchange coupling (which often appears in conjunction with the largest GMR values) through exact control of the thickness of the Cu spacer film.

Therefore, the goal of achieving a large GMR at a low field will probably best be reached through the development of improved techniques for the control of atomic structure during thin-film deposition. One avenue for such improvement that has not received much attention is deposition at lower substrate temperatures. Studies have been made at temperatures slightly below RT, such as 20 °C[4] and 0 °C,[6] but none far below RT. Studies of epitaxial growth have shown that deposition at low substrate temperatures can greatly modify film growth for metals such as those used in GMR films.[7,8] Phenomena such as surface diffusion, interdiffusion, surface segregation, agglomeration, etc., can often be suppressed or eliminated altogether by deposition at temperatures 100 °C or more below RT.[7,8] Thus, it would seem worthwhile to investigate the effects on GMR films of deposition at low substrate temperatures.

The present article can only be considered to be a preliminary investigation of this topic. In a GMR film with six different metal films, each of which might have its own optimum temperature for deposition, the number of samples required for a thorough investigation is prohibitively large. For example, deposition of a particular layer at a single temperature might not be ideal. Only the first monolayer (ML) or two of a given film might need to be deposited at low temperature (e.g., to suppress surface segregation of the underlying metal), and the remainder of that film might be better grown at a higher temperature.

Clearly, mapping out the ideal combinations of temperature, time, and thickness for each of the films will not be accomplished soon. Nevertheless, we have identified several important factors in spin-valve properties. We have identified the valleys that exist between grains in these polycrystalline films as an important form of roughness. These valleys impair spin-valve properties by producing the magnetostatic coupling field observed in these films. Suppressing the depth of these valleys by low-temperature deposition is desirable.

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Increasing their depth by deposition above RT gives an undesirable increase in coupling. Another factor we have identified (through analogies with single-crystal studies) is the interdiffusion that occurs when Co is deposited on Cu at RT or above. This interdiffusion appears to decrease the GMR and increases the coupling. This interdiffusion may be suppressed by low-temperature deposition. These insights and others described below represent a promising beginning to the fabrication of spin valves with improved properties.

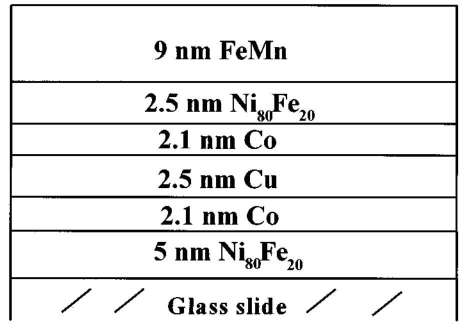


FIG. 1. An illustration of the standard spin-valve structure that is the basis for the present investigation.

## II. EXPERIMENT

The substrates used in this work were 12 mm diameter cover-glass slides, cleaned ultrasonically, rinsed in distilled water, dried, and installed in the deposition chamber. The base pressure before depositing a spin valve was typically $2\times10^{-8}$ Torr ($\sim10^{-6}$ Pa) of which ~95% was $H_2$ and the remainder largely $H_2O$. The presence of $H_2$ during deposition has no apparent effect on spin-valve properties unless the partial pressure exceeds $\sim10^{-6}$ Torr. The low base pressure is achieved partly by depositing a ~1.5 nm Ti film on the inside of the deposition chamber from a centrally mounted Ti filament just prior to deposition of each spin valve.

It is very important to remove the hydrocarbon contamination (several tenths of a nm of which is accumulated on the glass substrate from exposure to the laboratory air) prior to the deposition of each spin valve in order to achieve the highest GMR values. Substrates were sputtered with a neutralized-beam Ar ion gun at a beam energy of 500 eV until the carbon was removed, as judged by x-ray photoelectron spectroscopy (XPS) measurements in a connected vacuum chamber.

The metals films were deposited by dc-magnetron sputtering in 2 mTorr Ar at a rate of ~0.1 nm/s. During deposition, the samples were subject to an in-plane field of ~20 mT (200 Oe) provided by permanent magnets mounted on either side of the sample on two quartz–crystal-oscillator holders. The magnetoresistance measurements were made in the dc mode in another connected vacuum chamber using a four-point probe with a $5\frac{1}{2}$ digit ohm meter. Values of the four-point resistance can be converted into sheet resistance by multiplying by 4.1.

A scanning tunneling microscope (STM) is located in a separate chamber so that samples can be transferred through a vacuum interlock and characterized in vacuum. All images were recorded with a tunneling current of 0.2 nA with the tip biased at −50 mV with respect to the sample. The tips were prepared from 0.25 mm $Pt_{90}Ir_{10}$ wire clipped under tension with a wire cutter. For the STM data discussed here, multiple images were taken at a variety of locations on each sample to ensure that the results were typical. Care was taken to ensure that the results were not influenced by the use of different tunneling tips. Most STM images were recorded with a single tip, and great effort was devoted to repeated intercomparisons among the samples to ensure that changing tip conditions did not change the average roughness.

## III. RESULTS AND DISCUSSION

### A. The standard sample

The present work was based on a rather common type of spin valve structure, FeMn/$Ni_{80}Fe_{20}$/Co/Cu/Co/$Ni_{80}Fe_{20}$, which often achieves a moderate GMR at a rather low coercivity.[9] The top two magnetic films (Co and $Ni_{80}Fe_{20}$) are pinned by exchange bias from the FeMn, and the bottom two magnetic films are free to switch at low applied fields (unpinned). Adjacent Co and $Ni_{80}Fe_{20}$films are coupled so strongly that they always switch as a single magnetic unit.

The standard sample of this type used as a reference point in the present work is illustrated in Fig. 1. This standard sample was developed (and optimized for RT deposition) prior to any low-temperature studies.

Figure 2 presents the high- and low-field magnetoresistance data for a typical standard RT sample of this type. Figure 2(a) is the high-field data in which both pinned and unpinned films undergo switching. The switching of the unpinned films causes the loop observed near zero field, and the switching of the pinned films causes the loop observed around 14 mT. In the low-resistance state the magnetizations are parallel, and in the high-resistance state the magnetizations are antiparallel. The loop of the pinned films is shifted ~14 mT from zero field by the exchange bias from the FeMn.

Figure 2(b) presents the low-field data in which only the unpinned bottom two magnetic fields switch, while the magnetization of the top two magnetic films is held fixed (or pinned) by the FeMn. The center of the loop of the unpinned films is shifted 0.86 mT from zero field by the coupling field that exists between the top two magnetic films across the Cu (an offset in the positive field direction means the coupling field is ferromagnetic in sign).

The thicknesses indicated in Fig. 1 represent what we considered to be an optimum compromise between large values of the GMR and small values of the coercivity and coupling field. As an example of the trade-offs one faces in samples such as these, the GMR can be increased from 8% to about 9% by omitting the 5 nm $Ni_{80}Fe_{20}$ (and making the bottom Co film 5 nm thicker), but as a result the coercivity rises from less than 5 Oe to more than 20 Oe. It is necessary, in this case, to make the bottom Co 5 nm thicker when the 5 nm $Ni_{80}Fe_{20}$ is omitted because otherwise the pinning of the

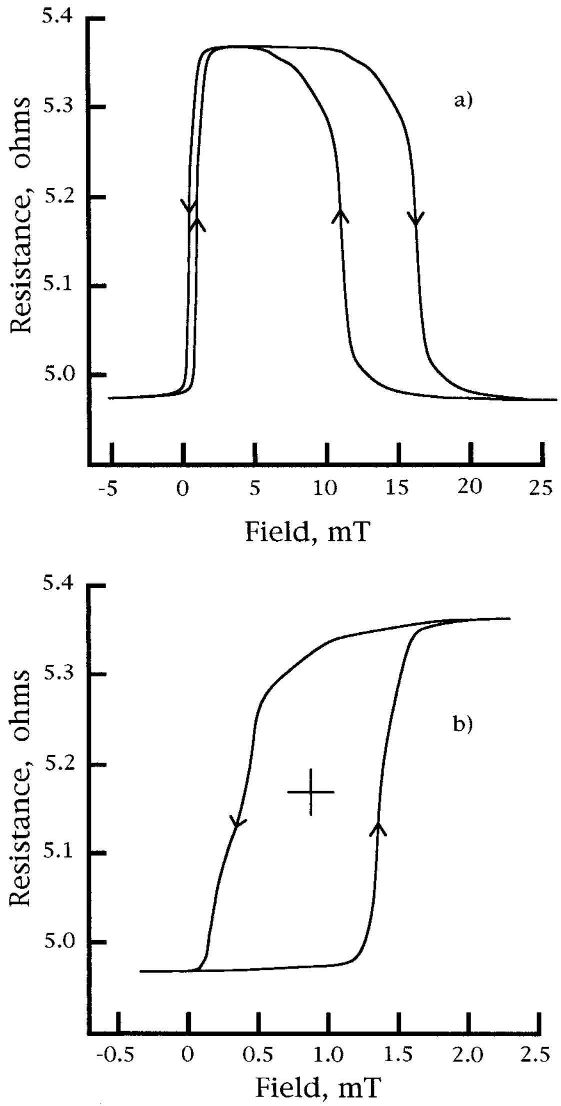


FIG. 2. The magnetoresistance loops for a sample of the type illustrated in Fig. 1 (RT deposition) for (a) The high-field case in which both pinned and unpinned magnetic films undergo switching and (b) The low-field case in which only the unpinned magnetic films undergo switching. The magnetic field is in units of milli Tesla (1 mT=10 Oe).

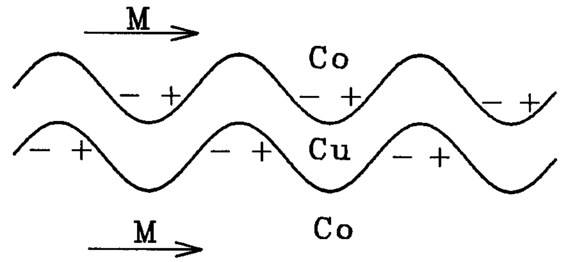


FIG. 3. An illustration of the orange peel coupling idea of Néel[10] in which magnetostatic coupling occurs due to the interaction of magnetic poles in a magnetic/nonmagnetic/magnetic structure with conformal roughness.

top magnetic films by the FeMn is too weak. This observation is one of the ironies we encountered in this work. The thickness of the bottom film (e.g., the 5 nm $Ni_{80}Fe_{20}$) affects the performance of the top film, five layers above it! From further studies we established that this phenomenon occurs because the strength of the pinning by FeMn depends strongly on the total thickness of the underlying metal films (but only weakly on how this total thickness is distributed among $Ni_{80}Fe_{20}$, Co, and Cu).

### B. The coupling field

In general, it is desirable to have as thin a Cu film as possible because thinner Cu generally increases the GMR. However, a lower limit is set by the coupling field which increases sharply below 2.5 nm Cu. For example, in our standard (RT) samples the coupling field is 0.86 mT (8.6 Oe) for 2.5 nm Cu (as in Fig. 2), but if we use 2.0 nm Cu the coupling field increases to 3.6 mT (36 Oe). This increase causes the pinned and unpinned loops to overlap partially so that the antiparallel state is never achieved completely, and the GMR actually decreases (to less than 5%) for 2.0 nm Cu.

A slight indication of such an overlap of the pinned and unpinned loops is apparent in Fig. 2(b), which presents the unpinned loop. It may be noticed that the loop is wider at the base than at the top. This characteristic occurs because in coming down from a high positive field the pinned film does not become completely saturated before the unpinned film begins to switch. In Fig. 2(b) this effect is barely noticeable, but is an indication that the 2.5 nm Cu film is the thinnest that is practical. This overlap becomes much more pronounced if the Cu film is even 0.2 nm thinner, and the GMR begins to decline.

For our standard (RT) samples of the type illustrated in Fig. 1, the coupling field is always ferromagnetic and is due primarily to the magnetostatic interaction across the Cu which follows from film roughness. The most important form of roughness is the long-wavelength roughness represented by the valleys between grains (as seen in the STM images). This long-wavelength roughness should, according to Néel's model,[10] make a major contribution to the coupling field. Figure 3 illustrates the Néel model. When two magnetic films are separated by a nonmagnetic film, any bumps or protrusions in the magnetic films will have magnetic poles on them, and a dipole fields will be set up (this model assumes the magnetization is in the plane of the film). If this roughness is conformal (i.e., if the same bumps occur in all three films one above another), then the dipole fields will interact in a manner that tends to produce parallel (or ferromagnetic) alignment in the magnetic films. This effect was termed "orange peel" coupling by Néel.[10] Our STM results generally confirm this concept, in that rougher samples tend to exhibit larger coupling fields than smoother ones, and the values of the coupling that we calculate using the Néel model and roughness data from the STM images is generally within about 20% of the observed values, which suggests that the coupling is indeed magnetostatic. Furthermore, the coupling fields for our standard (RT) samples are almost identical in measurements made at 150 K and at RT, as would be expected from a magnetostatic effect. The oscillatory coupling often found in GMR superlattices does not appear to play a noticeable role in our standard (RT) samples of the type illustrated in Fig. 1 [although it is significant for samples deposited at low temperature with thinner Cu (see below), where we found a much larger coupling strength at 150 K than at RT, as is normally found for the oscillatory effect].

Some simple principles may be inferred from Fig. 3. First, the steeper the slopes the greater will be the magnetic pole density and hence the stronger the coupling. Second, the coupling will be stronger when the peaks and valleys are closer to one another (e.g., smaller grain size). Finally, the coupling will be stronger for thinner Cu films.

### C. General considerations for low-temperature growth

For our low-temperature studies, 150 K was chosen because it is the lowest temperature that could be achieved reasonably quickly (within ~20 min). Since this temperature is approximately half RT, there should be a pronounced effect on thermally activated diffusion processes, which are exponentially dependent on sample temperature. For example, adatoms on (100) terraces of face-centered cubic (fcc) metals are highly mobile at RT for the metals used here, but at 150 K they are almost completely immobile on the time scale of our deposition.[7,8,11–13] In single-crystal studies, this room-temperature mobility typically gives rise to about two MLs of interdiffusion when a high-surface, free-energy metal such as Co is deposited on a low-surface, free-energy metal such as Cu.[7,8,14,15] Single-atom-high or monatomic steps are known to play an important role in initiating the interdiffusion, and although the mechanisms of this effect are not fully known yet, the evidence suggests that monatomic steps are constantly emitting and recapturing surface vacancies and adatoms on Cu surfaces at RT.[14] The interdiffusion processes may thus be viewed as involving place exchange of Co and Cu atoms and/or surface segregation of Cu during the deposition of the first ML or two of Co. It appears that such interdiffusion is largely suppressed at 150 K.[7,8]

Another factor to consider is that small irregular islands (a few atoms across) will be common on fcc(100) terraces after deposition of a given film at 150 K due to limited adatom mobility.[7] If the next film is deposited at 150 K its atoms will fill in the gaps to leave an intermixed ML, even without any active interdiffusion. These irregular islands can be suppressed by annealing each film after deposition, but the subsequent cool-downs to 150 K become time consuming and present an opportunity for the adsorption of background gases. In our experience with standard (RT) samples, the exposure of the sensitive interior films of a spin valve to the background gases (at a pressure of $\sim 5\times10^{-8}$ Torr of which ~95% was $H_2$ and the remainder mostly $H_2O$) for about an hour usually causes a noticeable loss of GMR (the length of time required for GMR loss suggests the $H_2$ and $H_2O$ are probably not responsible and implicates other trace gases).

Even without such pauses, GMR spin valves are somewhat contaminated during deposition. In our studies, XPS shows that our films exhibit typically $0.3\pm0.2$ ML of adsorbed oxygen atoms and sometimes ~0.1 ML of carbon atoms after deposition. A dc-magnetron gun acts as an electron flood gun and as a source of energetic Ar and metal atoms. When these species strike the chamber walls, atoms and molecules are desorbed and can condense on the film being deposited. Direct current magnetron sputtering is not a very clean environment even under the best circumstances. Fortunately, many potentially contaminating adatoms and molecules tend to float out to the surface of the film during deposition (even at cryogenic temperatures),[16] thus reducing the amount of bulk contamination.

The grains in the polycrystalline films of this work are expected to be almost randomly oriented and will thus have high-index crystal planes as surfaces. Such surfaces may be viewed as consisting of various combinations of fcc(100), (110),and (111) terraces a few atoms wide, separated by monatomic steps. Based on available single-crystals studies, surfaces such as these should, with their very high step densities, be very prone to interdiffusion when Co is deposited on Cu at RT.[6,7,8,14,15]

It is quite unlikely that the Co films in these multilayers have a strong hexagonal close packed (hcp) component. The initial deposition of 5 nm of an fcc alloy like $Ni_{80}Fe_{20}$ should initiate fcc grains. Since spin-valve multilayers generally exhibit approximately columnar growth[17] and epitaxy within each column (the lattice match is good in these systems), one would expect little hcp content. Generally, a strong hcp concentration occurs in GMR multilayers only when Co is the majority component.[18]

On fcc(111) terraces, the activation energy for surface diffusion is generally low and adatoms will retain some mobility even at 150 K.[12,13] Whenever such terraces are only a few atoms wide, deposited adatoms should have enough mobility to reach a monatomic step even at 150 K and to bond there. At RT, such steps appear to be important sites for interdiffusion, but interdiffusion at steps is probably suppressed at 150 K since it is likely to be a process of higher activation energy than adatom diffusion on fcc(111) surfaces.[12,13] Less is known about fcc(110) systems, but the available evidence suggests that similarities with fcc(111) and fcc(100) are likely.[7,13] However, for all three crystal surfaces, the available evidence comes from molecular beam epitaxy, which is a gentler form of deposition than magnetron sputtering. The energetic recoil of Ar atoms during sputtering and the energetic arrival of metal atoms can only be expected to increase interdiffusion at interfaces in these systems. Therefore, a ML or two (and possibly more) of interdiffusions is likely when Co is deposited on Cu at RT.

When Cu is deposited on Co, much less interdiffusion is expected since the relative surface-free energies oppose mixing.[7] Nevertheless, some mixing may occur for Cu on Co because of the impact of energetic Ar or metal atoms, but it is difficult to quantify how much mixing occurs, and it may well be negligible.

The question of whether interdiffusion increases or decreases the GMR in the Co/Cu system has been somewhat controversial with the initial evidence favoring an increase,[19] much subsequent evidence favoring a decrease,[20] and some evidence showing no effect.[21] Such controversies may have their root in the fact that it is probably difficult to change only one structural property of a complex multilayer system at a time to get a definitive answer to such a question. For example, a change in deposition conditions that increases

TABLE I. The GMR, sheet resistance, coercivity, and coupling field for samples of the type illustrated in Fig. 1 using different combinations of RT and 150 K deposition. The results of two samples are presented for each set of conditions. All data recorded at RT. Note: 1 Oe=0.1 mT.

| Deposition temperature | GMR | $R_{\square}$ ($\Omega$) | Coercivity (mT) | Coupling field (mT) |
|---|---|---|---|---|
| Entirely at RT | 7.7% | 20.5 | 0.45 | 0.85 |
| | 8.0% | 20.5 | 0.45 | 0.86 |
| Entirely at 150 K | 8.8% | 19.3 | 0.47 | 1.3 |
| | 8.8% | 18.5 | 0.49 | 1.5 |
| Only second Co at 150 K | 8.5% | 17.6 | 0.51 | 0.67 |
| | 8.5% | 18.5 | 0.47 | 0.88 |
| Only Cu and second Co at 150 K | 9.3% | 18.5 | 0.63 | 0.41 |
| | 8.5% | 18.5 | 0.50 | 0.32 |
| Only Cu at 150 K | 4.0% | 23.4 | ~0 | 3.4 |
| | 6.3% | 23.8 | ~0 | 2.4 |

interdiffusion might also increase grain size, and these changes might have opposite effects on GMR, so that whether GMR increases or decreases might well depend on a delicate balance between opposing influences. Therefore, caution is appropriate in evaluating any sweeping claim not supported by a comprehensive set of evidence. In the present work, we can provide plausible interpretations for the results in our particular type of spin valve but cannot prove that these interpretations apply to other GMR systems. One such interpretation is that interdiffusion decreases the GMR in our spin valves.

### D. Low-temperature growth of the standard GMR spin valve

Table I presents a representative sample of our data for low-temperature (LT) growth of the standard spin valve illustrated in Fig. 1. When the entire sample is deposited at 150 K, the GMR is clearly somewhat larger than for RT deposition. A suppression of interdiffusion when Co is deposited on Cu is the most likely explanation. Supporting this interpretation is the drop observed in the resistivity. Copper carries a large share of the current in these structures,[22] and any alloying (in the sense of interdiffusion) should reduce the thickness of pure Cu and increase the resistivity.

It is interesting how other properties change for LT deposition. The coercivity, a property generally thought to be highly sensitive to the defects in a film, changes only slightly. The property with the largest change is the coupling field. The increase seen here can be interpreted using our STM data (to be published separately[23]) which show a smaller mean grain size for LT deposition. In the Néel model, the coupling should increase if the roughness is unchanged and the grain size is smaller.[10] Our STM data show that the grain size is ~35% smaller but the roughness is only ~13% smaller for LT deposition compared to the corresponding quantities for RT deposition. (For RT deposition, the mean grain diameter after deposition of the Cu film is 9 nm and the mean depth of the valleys is 0.6 nm.) Note that all STM measurements had to be made at RT. We expect that the grain size probably does not change upon warming to RT, but the long-wavelength roughness probably increases as thermal diffusion allows the valleys to approach their equilibrium depth. This scenario of smaller grains and smoother surfaces for LT deposition might suggest opposing contributions to the coupling. Nevertheless, the dominant effect, apparently, is the smaller grain size which brings the valleys close and increases the dipolar coupling.

Additional support for the idea that interdiffusion occurs when Co is deposited on Cu at RT is found in the results for samples for which only the second Co film was deposited at 150 K. When this second Co film is deposited at RT one would expect interdiffusion to be large because the relative surface-free energies provide a large driving force for the surface segregation of Cu, and this segregation leads to an intermixed region at the interface. Since Co/Cu is the only interface in the spin-valve structure for which this driving force is large and since LT deposition tends to suppress this segregation, the samples for which only the second Co film was deposited at 150 K should resemble, in this regard, the samples deposited entirely at 150 K. Table I bears this idea out. For both types of films, the GMR is higher than that of the standard (RT) samples and the resistivity is lower, since interdiffusion is suppressed.

The coupling field of the samples for which only the second Co film was deposited at 150 K is about the same as that of the standard (RT) samples. This result is quite reasonable since it suggests that the grain size and roughness are determined by the RT deposition of the prior films.

When only the Cu and the second Co are deposited at LT, the data again suggest that interdiffusion at the Co/Cu interface is suppressed. As Table I shows, the GMR is high and the resistivity is low compared to the standard (RT) samples. However, for these samples there is an additional interesting effect. The coupling field is small as compared to either the standard (RT) samples or the ones with only the second Co film deposited at 150 K. This interesting result is also easy to explain. However, it should be noted first that it seems unlikely that a larger grain size is responsible for the sharp reduction in coupling field because the grain size should be largely set by the prior deposition of 5 nm of $Ni_{80}Fe_{20}$ and 2.1 nm of Co at RT. (Films such as these tend to exhibit somewhat columnar growth with the different films more or less epitaxial within a column.[17]) A more likely explanation is that the valleys between grains on the Cu surface are not as deep for LT deposition as for RT deposition because the Cu surface is not heated to RT before Co deposition. Surface diffusion is almost certainly required to produce these valleys and, to the extent surface diffusion is suppressed at 150 K, the valleys should be suppressed. The equilibrium depth of such valleys is well known to be determined by a balance of surface and interfacial tensions at the site of the grain boundary emerging from the surface.[24]

There is experimental support for the idea that surface diffusion allows these valleys to deepen. For example, if during the deposition of the standard (RT) sample, the sample is

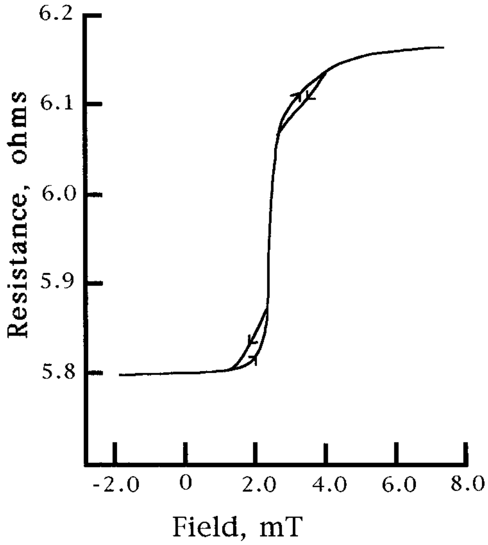


FIG. 4. The low-field magnetoresistance loop of a sample of the type illustrated in Fig. 1 for which the Cu was deposited at 150 K. The magnetic field is in units of mT (1 mT=10 Oe).

annealed for a few seconds at 200 °C after the deposition of Cu and is cooled to RT for deposition of the remaining films, the sample exhibits a coupling field over 3 mT (30 Oe), in contrast to the 0.85 mT (8.5 Oe) for the standard (RT) sample. The most likely explanation for this sharp increase is that the valleys deepen upon annealing because, as deposited, they are not at their equilibrium depth. One can only conclude that for LT deposition the valleys would be even shallower than for RT deposition.

Another mechanism for deepening of these valleys is negative substrate bias. We found that using a bias of −30 to −60 V during deposition causes a sharp increase in the coupling field. At bias values over −60 V, the coupling became too strong to observe any GMR. The impact of $Ar^+$ ions at these energies is well known to promote surface diffusion, much like the effect of annealing. (N.B., for substrate bias to have any effect at all, care must be taken to ensure that the sample surface has a good conducting path to the bias voltage.) This increased coupling is probably attributable to a deepening of the valleys. As an aside, we would not expect that ion-assisted deposition of these spin valves would yield improved results because of the similarity (in film bombardment effects) with negative substrate bias.

Additional insight into the complexity of LT deposition may be gained from examining one of its failures. It is clear from Table I that depositing only the Cu at LT is a failure. The GMR drops and the coupling rises sharply. Figure 4 presents the unpinned loop of such a sample. The coercivity appears to be zero, but it is probably being masked by a larger coupling field which dominates the switching, as explained below.

The likely explanation for the poor properties here is the condition of the Cu surface on which the Co is deposited. When the Cu film is deposited at 150 K and heated to RT (prior to the deposition of Co) the surface of the Cu film is only partially annealed. The many resulting small irregular islands provide a high density of monatomic steps (deposition at RT produces a much smaller step density than deposition at 150 K followed by warming to RT; see Ref. 11 for examples of this effect). As noted above, these steps may be expected to initiate severe interdiffusion when the Co is deposited on Cu at RT. If single-crystal studies may be used as a guide,[7,14,15] this interdiffusion should be much more extensive than that in the standard (RT) samples (for which the step density should be lower). The result of the greater interdiffusion is a reduction in the effective thickness of pure Cu. The resistivity of these samples is therefore high, as shown in Table I. As noted above, a reduction in Cu thickness generally increases the coupling field, and this reduction in turn increases the overlap in the loops and decreases the GMR. Both of these effects may be noted in Table I for these samples. Thus, an entirely plausible interpretation of the data is provided by the simple application of concepts well established in single-crystal studies.[7,14,15]

### E. Collapse of the loop by a large coupling field

The apparent low coercivity associated with a large coupling field in Fig. 4 is a common effect that we observe in GMR spin valves when the Cu film is thinner than optimum. The effect gives the appearance of a collapse of the loop and indicates that the coupling field does not act as a simple externally applied field (which would shift the position of the loop but not change its shape). The most likely explanation for this effect is that strong ferromagnetic coupling occurs in the vicinity of the valleys between grains. These valleys should, according to Néel's model,[10] make the major contribution to the coupling. However, this is a local effect. Therefore, the ferromagnetic magnetostatic coupling near such valleys should skew the magnetic moments in the bottom two (unpinned) magnetic films so that they turn somewhat into the direction of the top two (pinned) magnetic films even in the high-resistance state when the alignment is nominally antiferromagnetic. This effect helps to explain our general observation that the GMR begins to fall as the value of the coupling approaches the value of the coercivity. It also provides an explanation for the collapse of the loop. Apparently, the loop collapses when the coupling strength exceeds the coercivity because the switching mechanism changes from the motion of a domain wall in the sample to a local effect, the rotation of the moments under the dominating influence of strong coupling at the site of the valleys. Therefore, the loop takes on the appearance of a hard axis loop when the coupling is the dominant effect. This observation suggests that the term coupling ''field'' is a misnomer; the effect does not resemble that caused by a uniform externally applied field.

### F. Low-temperature growth and thinner Cu films

Given that LT deposition improves some of the properties of our standard spin valve, it is tempting to think that further improvements might be possible by combining LT deposition with changes in the formulation of the standard spin valve. Ideally, a complete reoptimization of all aspects of the standard spin valve would be appropriate. However, since this reoptimization would be very time consuming we

TABLE II. The GMR, sheet resistance, coercivity, and coupling field for samples of the type illustrated in Fig. 1 using thinner Cu films and two different deposition approaches (a) Deposition of the entire structure at 150 K and (b) deposition of only the Cu and second Co at 150 K. All data recorded at RT. Note: 1 mT=10 Oe.

| Cu thickness | GMR | $R_\square$ ($\Omega$) | Coercivity (mT) | Coupling field (mT) |
|---|---|---|---|---|
| (a) Deposition entirely at 150 K: | | | | |
| 2.5 nm | 8.8%, 8.8% | 19.3, 18.5 | 0.45, 0.47 | 1.3, 1.5 |
| 2.0 nm | 9.7% | 24.2 | 0.6 | 1.2 |
| 1.5 nm | 0.2% | 25.4 | ~0 | 12.0 |
| 1.0 nm | 11.4% | 30.0 | n.a. | −100 |
| (b) Only Cu and second Co at 150 K: | | | | |
| 2.5 nm | 9.3%, 8.5% | 18.5, 18.5 | 0.63, 0.50 | 0.41, 0.32 |
| 1.9 nm | 9.7% | 25.6 | ~0.2 | 2.1 |
| 1.5 nm | 0.2% | 28.3 | ~0.1 | 12.5 |
| 1.0 nm | 11.5% | 30.9 | n.a. | −100 |

have investigated the parameter most likely to produce interesting results, that of reducing the Cu thickness. Table II presents examples of our data.

The two cases for which thinner Cu is considered in Table II are the ones which gave the best results in Table I, deposition of the entire sample at 150 K and deposition of only Cu and the second Co at 150 K. Note that very poor results are obtained for the standard (RT) sample if the Cu is thinner than 2.5 nm. For example, the coupling is so large at a Cu thickness of 2.0 nm (3.6 mT or 36 Oe) that the loops overlap and the GMR is only 5%. No GMR is observed at all if the Cu is only slightly thinner.

Table II shows that dramatic improvements result from thinner Cu films for deposition entirely at 150 K [Table II(a)] or for deposition of only the Cu and the second Co film at 150 K [Table II(b)]. For example, the GMR rises to 9.7% with 2.0 nm or 1.9 nm or Cu and the couplings of 1.2 mT (12 Oe) and 2.1 mT (21 Oe) are significantly below the vale of 3.6 mT (36 Oe) that is found with 2.0 nm Cu in samples deposited entirely at RT.

Remarkably, Table II(a) shows that the coupling is less for 2.0 nm Cu than for 2.5 nm! This would be a very odd result, in view of the general increase in the coupling strength with decreasing Cu thickness, were it not for the results found with a 1 nm Cu film. The 1 nm Cu results in Table II indicate that the well-known oscillatory exchange coupling effect of Co/Cu superlattices[25] is present in these films. This presence accounts for the odd result. The oscillatory coupling exhibits an antiferromagnetic (AF) maximum at a Cu thickness of 2.2 nm.[25] At a Cu film thickness of 2.0 nm the magnetostatic (ferromagnetic) coupling should be strongly suppressed by this nearby AF maximum. This result is not only interesting, but in fact may be useful since it should be possible to bring the coupling very close to zero in such samples by slight adjustments in the Cu thickness.

The MR loops found for a 1 nm Cu thickness [Tables II(a) and II(b)] have a very different appearance from those shown in Fig. 2. Instead of the high- and low-field loops characteristic of simple spin valves, these samples exhibit bell-shaped curves centered near zero field that are characteristic of the well-known oscillatory exchange coupling effect of Co/Cu superlattices. These curves saturate at a field of about 100 mT (1000 Oe) at RT. If the sample is cooled to 150 K saturation occurs about 160 mT (1600 Oe), an increase that is characteristic of the oscillatory effect.[25] The minus sign in Table II indicates AF coupling. The energy of this coupling is 0.63 mJ/m$^2$ (0.63 erg/cm$^2$) at RT.

The achievement of AF exchange coupling for a 1 nm Cu film thickness is unprecedented for a simple spin-valve structure and is probably a consequence of LT deposition that suppresses interdiffusion and reduces the depth of the valleys. Moreover, the coupling energy is over four times larger than the 0.15 mJ/m$^2$ (0.15 erg/cm$^2$) found in Co/Cu superlattices,[25] even though the coupling in a simple spin valve comes only from one side of each Co film rather than from two sides as in a superlattice. Thus, the intrinsic coupling strength is over eight times larger!

Normally (i.e., for RT deposition), only superlattices have the necessary degree of structural perfection to exhibit AF coupling for a 1 nm Cu film thickness.[9] It is not known what type of structural perfection permits AF coupling in superlattices but prevents it in simple spin valves (for a 1 nm Cu film thickness and RT deposition), but it seems likely that it is related to grain size, and the resulting distance between valleys. The oscillatory AF coupling is a delicate effect easily destroyed by imperfections, and the typical depth of the valleys after Cu deposition in our spin valves for RT deposition is 0.6 nm, which is not much less than the 1 nm thickness of the Cu. Thus, it is understandable that AF coupling is never found for simple spin valves deposited at RT using a Cu thickness of 1 nm. Indeed, even GMR has not previously been reported for simple spin valves at a Cu thickness of 1 nm. Although this lack of GMR is generally blamed on ''pinholes'' in the Cu (where the upper Co is presumed to contact the lower Co), it seems more likely that the strong magnetostatic coupling at the valleys is responsible.

The probable reason for superlattices exhibiting AF coupling with RT deposition is that they are much thicker, and there is generally a steady increase in grain size with film thickness since some grains die out at the expense of others that grow wider (the growth is only approximately columnar).[17] Thus, the superlattices should have a lower density of valleys to perturb the AF coupling.

The small GMR of samples with a Cu film thickness of 1.5 nm in Table II is not surprising. For this Cu thickness, no AF coupling is expected since the 1.5 nm thickness is midway between the AF maxima at 0.9 and 2.2 nm Cu.[21] Thus, there is nothing to offset the expected magnetostatic ferromagnetic coupling and, indeed, the oscillatory coupling with 1.5 nm Cu may well be ferromagnetic[26] and add to the strength of the magnetostatic ferromagnetic coupling. Thus, the large ferromagnetic coupling of ⩾12 mT (⩾120 Oe) is expected. The GMR is small in this case because the loops overlap severely.

The values of the sheet resistance in Table II show the expected monotonic increase with decreasing Cu thickness. There do not seem to be any important implications here.

It is a little surprising that the GMR is only about 11.5% for 1 nm Cu thickness (Table II). If Co/Cu superlattices may be used as a guide,[25,27] an increase in GMR to about 15% might have been expected at 1 nm Cu on the basis of the 9.7% result at 2 nm. The most likely explanation is that the magnetic films do not exhibit perfect AF alignment for a 1 nm Cu thickness. The obvious cause is likely to be the valleys where a structural perturbation impairing the oscillatory AF coupling would be most likely and where the magnetostatic, ferromagnetic coupling would be strongest. Thus, the direction of the two magnetizations probably twist out of perfect AF alignment at the valleys. Since the grain diameter in these films is smaller than the width of a typical domain wall in Co or $Ni_{80}Fe_{20}$, only a partial twisting out of AF alignment is expected. An effect somewhat like magnetization ripple might be envisioned for the unpinned films.

If the above interpretation is correct, thinner films of Cu should be possible without excessive coupling if better methods to suppress the valleys can be found. As a result, larger GMR values should be attainable. One possible route to this goal would be deposition at even lower temperatures than 150 K. The use of sputtering gases heavier than Ar (to suppress energetic recoil) is another. Still another approach would be the use of surfactant layers such as Pb or In, an approach we are currently pursuing. As mentioned above, one approach that fails badly is negative substrate bias during deposition.

## IV. CONCLUSIONS

The major conclusions of this work may be summarized as follows:

(1) The interdiffusion which occurs when Co is deposited on Cu at room temperature reduces the GMR of simple spin valves.
(2) Deposition at 150 K tends to suppress this interdiffusion and increases the GMR with little change in coercivity. As a result, GMR values of 8.8% and coercivities below 0.5 mT (5 Oe) are achieved for spin valves deposited entirely at 150 K.
(3) The interdiffusion reduces the effective thickness of the pure Cu film and thus increases the resistivity of the spin valve.
(4) The surface roughness, in the form of valleys separating the polycrystalline grains, is largely responsible for the ferromagnetic, magnetostatic coupling between the magnetic films across the Cu.
(5) This type of roughness can be reduced by deposition at 150 K, and if the grain size is unchanged, the coupling is likewise reduced.
(6) This coupling does not act as a simple externally applied field shifting an otherwise unchanged hysteresis loop. Instead, it alters the switching mechanism and collapses the loop whenever the coupling is significantly larger than the coercivity.
(7) When the entire spin valve is deposited at 150 K, the grain size is substantially smaller and the coupling is larger due to the increased density of valleys. As a result, the best combination of low coercivity, low coupling, and large GMR are found only when the Cu and the second Co films are deposited at 150 K.
(8) Deposition of spin valves above room temperature or with negative substrate bias yields poor results as the coupling becomes much larger. In these cases, the increased mobility of surface atoms probably accelerates the interdiffusion and increases the depth of the valleys.
(9) The improved quality of the interfaces due to deposition at 150 K allows the first maximum, at 1 nm Cu, in the oscillatory antiferromagnetic coupling (so common in Co/Cu superlattices) to be observed in simple spin valves for the first time.

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