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Interplay between oscillatory exchange coupling and coercivities in giant magnetoresistive [$Ni_{80}Fe_{20}$/Cu/Co/Cu] multilayers

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We have observed giant magnetoresistance in [$Ni_{80}Fe_{20}$/Cu/Co/Cu] multilayers with well-defined oscillations in magnitude as a function of the Cu layer thickness both at 300 and 4.2 K. The phase and period are found to be very similar to those previously measured in Co/Cu and, more recently, $Ni_{81}Fe_{19}$/Cu multilayers. However, the existence of a strong contrast in coercive fields between $Ni_{80}Fe_{20}$ and Co leads to a significant magnetoresistance for Cu layer thicknesses where coupling is zero or even moderately ferromagnetic. At room temperature, resistance changes as large as 7% are observed within a few tens of oersted of zero field.

Following the discovery a few years ago of a giant magnetoresistance in antiferromagnetically coupled Fe/Cr superlattices,[1] there have been a great number of works related to the transport properties of multilayers in which ferromagnetic and nonferromagnetic layers alternate. In fact, saturation magnetoresistances as large as 65% at room temperature have been found in sputter-deposited Co/Cu multilayers.[2] This is about 25 times larger than in permalloy thin films currently used in MR devices. Magnetic multilayers therefore represent a potential breakthrough as superior materials for technological applications such as MR read heads or magnetic sensors. However, in order to get a significant gain in terms of *sensitivity* (percentage of resistivity change per unit field near zero), it is necessary to produce large resistance changes with low applied fields. This challenge remains largely open.

More recently, there have been reports on multilayer (ML) structures which, in contrast with Fe/Cr or Co/Cu systems, do *not* depend on an antiferromagnetic (AF) interlayer exchange coupling to drive the moments into the antiparallel arrangement which establishes the so-called spin-valve effect. At room temperature, exchange-biased [NiFe/Cu/NiFe/FeMn] bilayers[3] were observed to display resistance changes of 3% to 4% within a field range of 5–10 Oe, while uncoupled [NiFe/Cu/Co/Cu] MLs[4] showed resistance changes of about 9% in a 100 Oe range. A much sharper field response can be expected in these latter structures considering the very low coercivities and saturation fields of isolated Permalloy thin films. On the other hand, an oscillatory interlayer exchange coupling has already been observed in both Co/Cu[2] and NiFe/Cu[5] MLs, and one might expect the same phenomenon in [NiFe/Cu/Co/Cu] in the proper Cu thickness range. If present, such an interlayer coupling would be of interest from a fundamental perspective, but would also be of prime practical interest because of the possible interplay between coupling and coercivities in determining the saturation field for the giant magnetoresistance.

Here, we report the observation of well-defined oscillations in the saturation values of the MR as a function of Cu layer thickness in [$Ni_{80}Fe_{20}$/Cu/Co/Cu] multilayers. The phase and period found are similar to those of both Co/Cu[2] and NiFe/Cu[5] MLs. Furthermore, we observe the crossover from a magnetization process dominated by exchange coupling, for Cu layer thickness below 20–30 Å, to an uncoupled behavior at large thicknesses which is mainly controlled by the differences in coercivities, as described in Ref. 4.

The MLs were prepared by rf diode sputtering in a Nordiko NS 2000 high-vacuum computerized sputter deposition system. Prior to deposition, the base pressure was in the low 10^{-7} Torr range. The films were deposited at room temperature in an Ar pressure of 5 mTorr at deposition rates of $\sim$0.5–1 Å/s, depending on the material. We used chemically etched, highly resistive Si(100) wafers as substrates. The layer thicknesses were determined from deposition times following the calibration of the deposition rates using a DEKTAK profilometer. In order to improve the overall quality of the ML, we always begin the growth by depositing a 60 Å iron buffer layer on the Si.[2]

The structure of the MLs were assessed on selected samples by transmission electron microscopy (TEM) and x-ray diffraction (XRD). Figure 1 shows a TEM cross section under suitable viewing conditions (strong underfocusing) for a typical ML. The multilayer stacking is clearly visible. Near the substrate, layers appear to be almost flat, with roughness below 10 Å. After three periods, a degradation of the flatness is noticed and, at the top of the ML, a waviness with an amplitude in the 50 Å range is observed. However, this waviness does not affect the ML stacking, each layer being apparently continuous on a large scale. Our objective being the careful study of the influence of exchange coupling on the properties of [NiFe/Cu/Co/Cu] MLs, we limited the number of periods to three in the sample set considered here in order to avoid any uncontrolled effects related to the waviness in the ML structure. The results reported below correspond, therefore, to the series of samples described by [50 Å NiFe/t_{Cu} Å Cu/20 Å Co/t_{Cu} Å Cu]$\times$3, where t_{Cu} is the thickness of the copper layer.

Dark-field TEM measurements indicate that the crys-

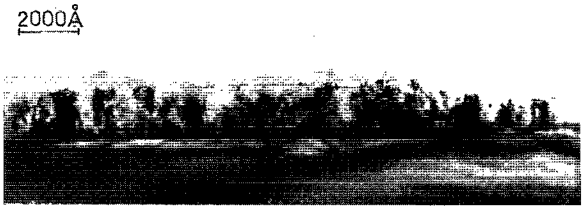

FIG. 1. TEM cross section for [50 Å NiFe/50 Å Cu/10 Å Co/50 Å Cu]$\times$12 multilayer structure.

tallites show a columnar shape and may propagate through the whole thickness of the ML. This indicates that a significant part of the interfaces between the layers are coherent from a crystallographic point of view within a grain. Furthermore, dark-field observations of the diffuse scattering reveal that, at the interface between the Fe buffer layer and the Si substrate, there is a 15 Å amorphous layer reflecting a possible formation of iron silicide at the interface and/or residual SiO_x presence.

The ML structure is polycrystalline as revealed by XRD and electron diffraction. The grain size, measured on plane views in TEM, ranges between 200 and 1000 Å. XRD also indicates that most of the structure is fcc despite a residual hcp contribution. XRD and electron diffraction show some [111] texture on these MLs. All these observations are very similar to previously reported results[2] on Co/Cu.

Magnetization measurements were performed at room temperature on 5×10 mm^2 pieces cut from the Si wafers using a vibrating sample magnetometer. On similar unpatterned samples, we performed the magnetoresistance measurements using a standard dc four-point technique. The relative changes in resistance were calculated as usual using the high-field resistance as a reference.

Both the hysteresis loops and the *R-H* curves show qualitative variations with spacer thickness t_{Cu} corresponding to the passage through AF-coupled, uncoupled, and F-coupled regimes. This is illustrated in Figs. 2 and 3 which show room-temperature *M-H* and *R-H* cycles, respectively, for two samples. The AF interlayer coupling is clearly visible in the data from the sample with $t_{Cu}=10$ Å. The hysteresis loop for this sample shows a high saturation field of about 1 kOe which is much larger than the coercivities of the component layers. The *R-H* cycle for the same sample displays a broad maximum near zero, with an almost complete overlap of ascending and descending branches, such as those observed in other AF-coupled multilayers.[1,2] Although a significant remanent magnetization is observed in the hysteresis loop, this is consistent with the presence of the relatively thick 60 Å iron buffer which is in direct contact with the first permalloy layer. In fact, assuming all component layers have the nominal thicknesses and bulk saturation magnetizations, one expects a remanence of 44% for a perfectly antiparallel or "giant ferrimagnetic"[4] configuration of the NiFe and Co component layers. The observed value of 50% actually shows that we are very close to that state.

Figures 2(b) and 3(b) correspond to the sample with

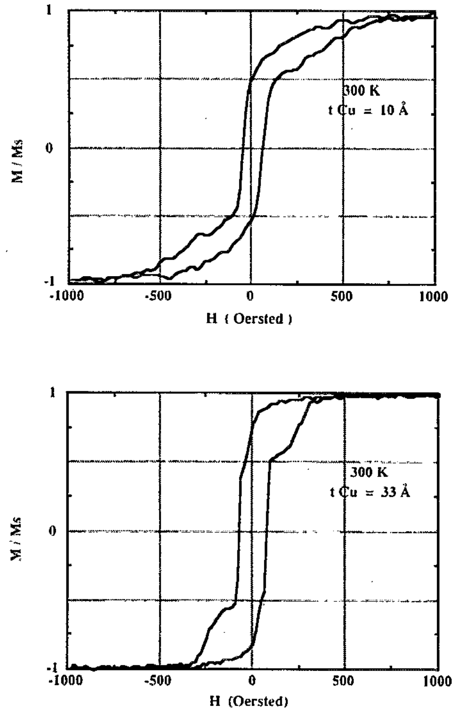

FIG. 2. Hysteresis loops for two samples with $t_{Cu}=10$ and 34 Å. The first sample shows an AF interlayer coupling, while the second one displays a nearly independent switching of NiFe and Co magnetizations.

$t_{Cu}=34$ Å. The hysteresis loop displays the two-step reversal process which has previously been observed in this system.[4] The *R-H* curve is also characteristic of MLs with two types of magnetic component layers, featuring asymmetric maxima with a quasiplateau in the field range between the contrasting NiFe and Co coercivities. There is almost no overlap of the peaks for the ascending and descending branches. The samples with t_{Cu} of 6 and 16 Å display hysteresis loops which are quite square compared to those shown here, and *R-H* curves which are comparatively flat. This is consistent with a *ferromagnetic* interlayer coupling for those thicknesses.

The overall variation of the MR with t_{Cu} is displayed in Fig. 4. Three distinct peaks can be seen near $t_{Cu}=10$, 22, and 34 Å. The period of about 12 Å and the position of the first peak is similar to what has been observed in Co/Cu and NiFe/Cu multilayers.[2,5] This similarity supports the current interpretation of the oscillatory interlayer exchange coupling, according to which the oscillation period depends primarily on the Fermi surface and crystal structure of the spacer layer.[6]

Aside from the similar period, the MR versus t_{Cu} oscillations displayed in Fig. 4 differ somewhat from those observed in Co/Cu and NiFe/Cu systems. Consider imaginary two "envelope" curves joining the maxima and minima, respectively. For all three of these systems, the envelopes have virtually converged after three oscillations. In NiFe/Cu and Co/Cu, the "converged" MR is a small fraction of value at the first peak. For the present system, however, the curve joining the minima rises to meet the

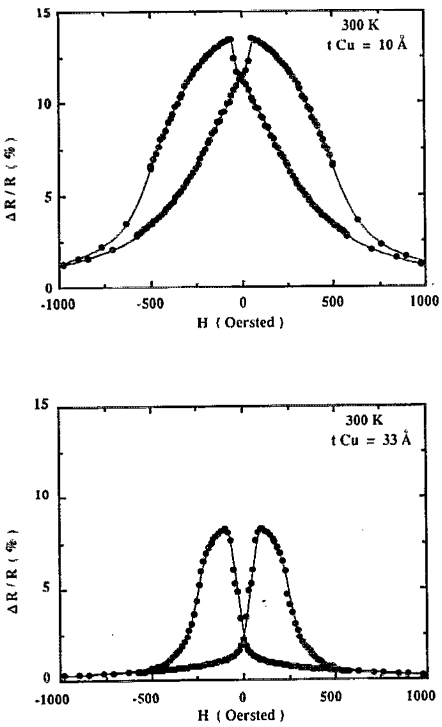

FIG. 3. MR curves ($\Delta R/R$ vs H) for two samples with $t_{Cu}=10$ and 34 Å. The passage from a significant AF coupling to a regime dominated by contrasting coercivities is visible in the relative values of maximum and zero-field resistance.

envelope of the maxima, and the MR which remains after the oscillations have died out is similar in magnitude to the first peak. This situation does not change when a correction is made for the shunting effect of the iron buffer layer.

This unique dependence of the MR on t_{Cu} is thus intimately related to the coexistence, in [NiFe/Cu/Co/Cu] multilayers, of *two* mechanisms which affect the degree of antiparallel ordering achieved in low fields: the interlayer exchange coupling and, when the former is weak, the contrasting coercivities of NiFe and Co component layers. Even when the coupling has become insignificant, a nearly antiparallel configuration is reached during the magnetization reversal process. The decrease in maximum MR values between consecutive peaks in Fig. 4 is due primarily to the spin-valve effect's intrinsic decrease with t_{Cu}.[7] This stands in contrast to the variation of the peak heights with spacer thickness in ML systems such as Co/Cu, where an intrinsic decrease of the MR with t_{Cu} is accompanied by a strong reduction in the strength of the interlayer coupling. The uncoupled films never achieve a well-defined antiparallel configuration, so the MR measured at a large t_{Cu} corresponds to an incomplete or "partial" spin-valve effect. As a consequence, there is an enhancement of the MR's dependence on spacer thickness.

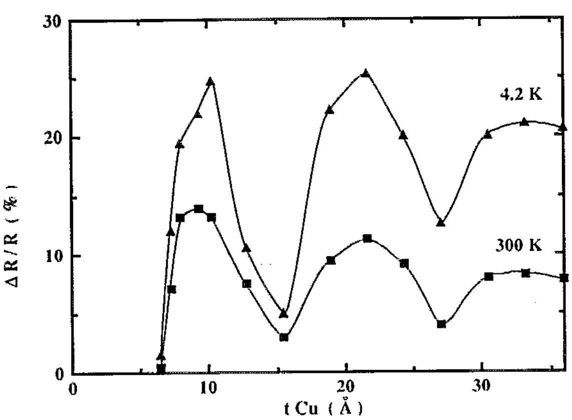

FIG. 4. Variation of the MR with spacer thickness t_{Cu} at 4.2 and 300 K. The solid lines are to guide the eye.

In conclusion, we have presented results showing that an oscillatory interlayer exchange coupling is present in [NiFe/Cu/Co/Cu] multilayers. The oscillation period and phase were found to be similar to what has been observed in NiFe/Cu and Co/Cu multilayers. The nature of the oscillations was found to differ from what has been seen in other systems due to the interplay between interlayer coupling, which is dominant when t_{Cu} is small, and the contrasting coercivities of the NiFe and Co component layers, which become manifest in the weakly coupled regime. The coexistence of these two mechanisms needs to be taken into account in considering the possible technological applications, and could provide strategic additional control over the giant magnetoresistance in this system. For this series of samples, a maximum sensitivity of about 0.17%/Oe at room temperature was obtained between zero and 40 Oe for the sample with $t_{Cu}=26$ Å, which is near the *second* AF peak.

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